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Mid-late Holocene environmental and cultural dynamics at the south-west tip of Europe (Doñana National Park, SW Iberia, Spain)

José Antonio López-Sáez[a,*], Sebastián Pérez-Díaz[a], Antonio Rodríguez-Ramírez[b], Antonio Blanco-González[c], Juan J.R. Villarías-Robles[d], Reyes Luelmo-Lautenschlaeger[a], Gonzalo Jiménez-Moreno[e], Sebastián Celestino-Pérez[f], Enrique Cerrillo-Cuenca[f], José N. Pérez-Asensio[g], Ángel León[h]

[a] G.I. Arqueobiología, Instituto de Historia (CCHS), C.S.I.C., Albasanz 26-28, 28037 Madrid, Spain
[b] Departamento de Geología, Universidad de Huelva, Avenida de las Fuerzas Armadas s/n, 21007 Huelva, Spain
[c] GIR PREHUSAL, Universidad de Salamanca, Cervantes s/n, 3700 Salamanca, Spain
[d] Instituto de Lengua, Literatura y Antropología (CCHS), C.S.I.C., Albasanz 26-28, 28037 Madrid, Spain
[e] Departamento de Estratigrafía y Paleontología, Facultad de Ciencias, Universidad de Granada, 18071 Granada, Spain
[f] Instituto de Arqueología de Mérida, C.S.I.C., Plaza de España 15, 06800 Mérida, Spain
[g] GRC Geociències Marines, Departament de Dinàmica de la Terra i de l'Oceà, Facultat de Ciències de la Terra, Universitat de Barcelona, Martí i Franques s/n, 08028 Barcelona, Spain
[h] Fundación del Hogar del Empleado (FUHEM), Colegio Lourdes, San Roberto 8, 28011 Madrid, Spain

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ABSTRACT

A multi-proxy palaeoenvironmental study (pollen, non-pollen palynomorphs, charcoal particles, mollusk macrofauna) of coastal marshland in Doñana National Park (southwestern Iberian Peninsula) was undertaken to trace environmental change, human activities related to woodland clearance, and past land-use during the mid-late Holocene (~5000–2800 cal BP). The results of this study are combined with archaeological data from the Copper and Bronze Ages and are subsequently compared with those of similar research carried out at the south-westernmost part of Europe with the aim of discerning regional differences or similarities. Our research has allowed us to recognize climate changes and four extreme wave events in the Guadalquivir paleoestuary, which might have contributed to both the cultural change that is observed in the archaeological record at the end of the Chalcolithic and the subsequent population decline during much of the Bronze Age.

1. Introduction

During the Upper Pleistocene and the Holocene, the south-western extreme of Europe is a territory of major archaeological and anthropological interest due to its border position at the crossroads between Europe and Africa and between the Atlantic Ocean and the Mediterranean Sea (López-García and López-Sáez, 1994a, 1994b; Pérez-Díaz et al., 2017). In the southwest of the Iberian Peninsula it has been documented the earliest known use of marine resources was by Neanderthals ~150 kyr ago (Cortés-Sánchez et al., 2011) and was a crucial reservoir of biodiversity during the Upper Pleistocene and early Holocene (Carrión et al., 2008; Cortés-Sánchez et al., 2008). Southwest Iberia is also key to understanding the neolithization process in the Iberian Peninsula; evidence of Neolithic settlement is present from at least 7500 cal BP, when an agricultural and food producing economy quickly replaced the coastal economies of the Mesolithic populations after the 8200 cal BP abrupt climatic event (López-Sáez et al., 2011; Cortés-Sánchez et al., 2012). Southwest Iberia is a unique area for the study of the funerary record and demography from the Mesolithic to the Copper Age (Díaz-Zorita et al., 2012), when some highly-elaborate tombs, such as Montelirio (Seville), feature ceremonial and rich burial goods unparalleled in Chalcolithic Europe (Fernández-Flores et al., 2016). Importantly, this region exhibits sharp cultural disruptions at the time of the 4200 cal BP climatic event and is, therefore, crucial for tracking plausible population movements between the SW and the SE of the Iberian Peninsula (Lillios et al., 2016; Blanco-González et al., 2018).

* Corresponding author.
E-mail addresses: joseantonio.lopez@cchs.csic.es (J.A. López-Sáez), sebastian.perez@cchs.csic.es (S. Pérez-Díaz), arodri@uhu.es (A. Rodríguez-Ramírez), ablancoglez@usal.es (A. Blanco-González), juanjose.villarias@cchs.csic.es (J.J.R. Villarías-Robles), reyes.luelmo@cchs.csic.es (R. Luelmo-Lautenschlaeger), gonzaloj@ugr.es (G. Jiménez-Moreno), scelestino@iam.csic.es (S. Celestino-Pérez), jn.perezasensio@ub.edu (J.N. Pérez-Asensio).

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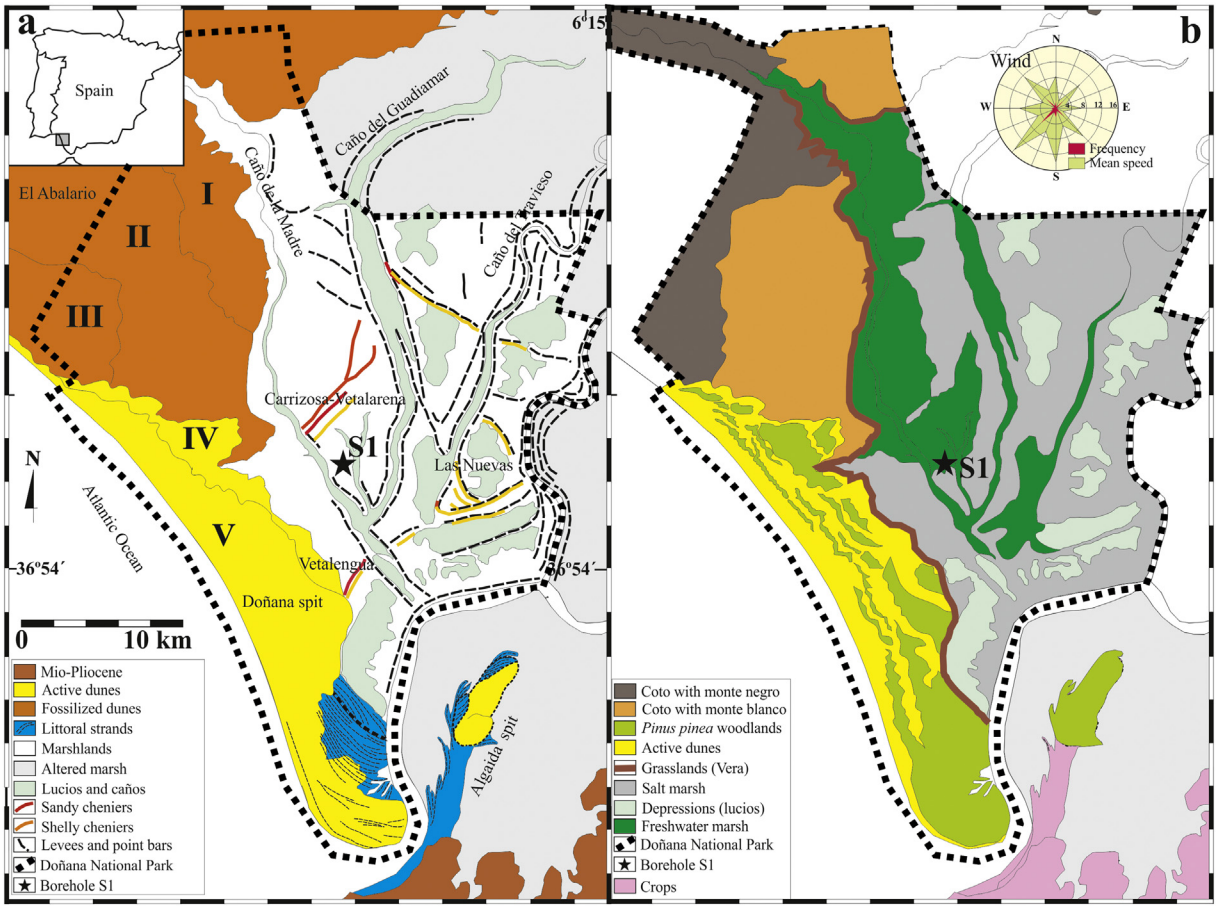

Fig. 1. Study area and location of the S1 core. (a) Geomorphological units of the Doñana National Park. (b) Vegetation map of the Doñana National Park. Wind directions are also indicated.

Finally, the timing and forms of interaction between local Late Bronze Age and Early Iron Age societies and the expansion of Mycenaean, Phoenician, and Greek influence, as evidenced by artefacts, are the subject of intense debate (López-Sáez et al., 2002b; Celestino-Pérez et al., 2008).

Within Southwest Iberia (Fig. 1), Doñana National Park (DNP) is one of the largest in Spain, the landscape formed as a result of a long and complex geological evolution (~6 Myr) and anthropogenic history (Celestino-Pérez et al., 2016). The area encompasses a large system of rivers, marshes, dunes, and beaches associated with coastal dynamics at the mouth of the Guadalquivir River. The archaeological literature regarding the formation of this landscape in the course of the Holocene is still under the influence of an uniformitarian model that Schulten (1945) and Gavala (1927, 1936) proposed in the first half of the 20th century, before the scientific impact of Plate Tectonics Theory. According to this model, the maximum transgression of the Atlantic Ocean during the Holocene opened up a wide estuary that extended as far inland as Coria del Río, some 15 km south of Seville. Thereafter, estuarine and riverine sedimentation began to infill the feature, leading to the development of a major delta and the progradation, of coastal barriers. Behind these barriers, a large inland lake developed, the *Lacus Ligustinus* mentioned by Rufus Festus Avienus in the 4th century AD (Rodríguez-Ramírez et al., 2016). Eventually, the lake was infilled, turning the landscape into a series of marshes and lagoons that anyone visiting the DNP today would recognize.

Elsewhere with in the DNP, large tracts of the former estuary have been reclaimed by constructing flood levees and undertaking large-scale agricultural projects. Consequently, it was not until this process of extensive infilling reached a certain level, not before the Middle Ages, that the area could have carried a significant human population. Later versions of such an uniformitarian view are, for instance, those of Menanteau (1981), Arteaga and Roos (1995), and Escacena (2001, 2014).

However, recent geological, archaeological, and paleoenvironmental research (Rodríguez-Ramírez et al., 2014, 2015; Jiménez-Moreno et al., 2015), including the research reported in this paper, point instead to a far more complex evolution. In this new model, geomorphological developments in the Guadalquivir paleoestuary in the Holocene, such as the formation of rivers, marshes, dunes, and beaches have been the result of complex interactions between natural processes (climate fluctuations, fluvial and coastal dynamics, and neotectonic movements) and human activities. By the end of the 5th millennium cal BP, for instance, much of the original paleoestuary had turned into marshland by river sedimentation into the paleoestuary, reducing it to a relatively small brackish lagoon; an extreme wave event (EWE) occurring ~4200 cal BP led to erosion and reversed this process, ushering in a new cycle of infilling in the paleoestuary (Rodríguez-Ramírez et al., 2015). Because of this long, complex history of natural and human evolution, DNP has been characterized as a coupled socio-ecological system covering four major ecosystem types: marshland, dunes, coastal and estuar (Ojeda, 1990; Gómez-Baggethum et al., 2010, 2012; Palomo et al., 2014). This socio-ecosystem depends on freshwater flows to sustain biodiversity and agriculture, making it extremely vulnerable to human or climate-induced changes in freshwater availability (Fernández et al., 2010; Martín-López et al., 2011). DNP is recognized as one of the most emblematic natural wetlands in Europe (Fernández-Delgado, 2006; Fernández et al., 2010). Its cultural and ecological importance is recognized through its declaration as a Ramsar site, a

UNESCO World Heritage Site, and as a Biosphere Reserve. Its relevance is also acknowledged through different conservation strategies covering about 1080 km^2, including a Natural Park as well as the National Park (García-Novo and Marín-Cabrera, 2006). Given the uniqueness of these environmental conditions within Southwest Iberia, and as a result of the high level of protection afforded to Doñana, Finlayson et al. (2008) consider DNP as one of the few remaining zones in Iberia that can be used as witness to the habitats and landscapes of the last Neanderthal populations. Furthermore, although Southwest Iberia is classified as a low-probability tsunamigenic area, at least sixteen EWEs, tsunamis and/or severe coastal storms have been identified so far between 7000 cal BP and 1900 CE. Five to seven of them are recorded as palaeotsunamis even though the palaeoenvironmental consequences of these events are poorly known (Ruiz et al., 2010; Lario et al., 2010, 2011; Morales et al., 2011; Rodríguez-Ramírez et al., 2015).

Since 2005, an integrated research programme involving geologists, palaeoenvironmentalists, archaeologists, historians, and anthropologists has been undertaken in DNP, with the aim of reconstructing the long-term landscape development and socio-ecological, cultural history of this coastal environment. Thus far, the programme has focused on understanding the complex, neotectonic - as well as climate-related geomorphological evolution of the Guadalquivir paleoestuary during the Holocene (Rodríguez-Ramírez et al., 2014) and on assessing the impact of EWEs on paleogeographical changes and human settlement in prehistoric, early historical, and Roman times (Rodríguez-Ramírez et al., 2015, 2016). The programme has also modelled the relationships between late Holocene climate variability and vegetation dynamics (Jiménez-Moreno et al., 2015) to provide insights into the geological, archaeological, and anthropological factors that have sustained the preservation of this singular natural space (Celestino-Pérez et al., 2016).

In this paper, we now present a record of mid-late Holocene (~5000–2800 cal BP) landscape change based upon the analysis of pollen, non-pollen palynomorphs, and macrocharcoal sampled from a core (S1) drilled in the marsh of Hinojos (DNP). The study aims to redress the research gap concerning the relationship between vegetation dynamics, human impact, and climate variability, and to develop a complete picture of the complexity of human activities around this coastal wetland landscape. We combine our new palaeoenvironmental data with those from other studies carried out in Southwest Iberia. The specific aims of this combined analysis were to: i) identify different human activities; ii) evaluate the strategies followed during different periods; iii) describe the marshland landscape response to human disturbance on a micro-regional scale; and iv) establish the causal relationships between potential tsunamis and other EWEs and the development of the marshland ecosystems of the Doñana National Park.

2. Study area

2.1. Geology and present-day geomorphology

The Iberian Peninsula lies at the southwestern corner of the continental component of the Eurasian plate, right across the northern boundary of the African plate, an area also known as the Azores-Gibraltar fault zone. The present geological structure of the Gulf of Cadiz is the result of the European-African plate convergence motion, dextral strike-slip along the Azores-Gibraltar Plate Boundary (Medialdea et al., 2009). The Gulf of Cadiz includes a number of estuaries containing deltas, all of them partly enclosed by coastal barriers or spits. The largest delta is, by far, that of the Guadalquivir River, enclosed by two spits: Doñana and La Algaida (Fig. 1a). The entire area comprises one of the largest wetlands (54,720 ha) in Europe and incorporates the Doñana National Park (DNP). The park lies on the right bank of the Guadalquivir River and extends over 50,700 ha along the coastal plain of the Gulf of Cadiz (Fig. 1a); it includes four morphogenetic systems: estuarine, fluvial, littoral, and aeolian (Rodríguez-Ramírez et al., 1996). The present-day landscape is the outcome of the complex geological evolution of the lower Guadalquivir River valley since late Miocene times, though the majority of recent change has occurred since the Last Glacial Maximum. Postglacial sea-level rise reached its maximum ~6500 cal BP and caused the drowning of the area (Zazo et al., 1994); since then, the Guadalquivir estuary has been enlarged and reshaped by the formation of sandy spits after the last postglacial transgression, littoral dynamics largely controlling such development in the present landscape (Rodríguez-Ramírez et al., 1996).

The growth of littoral spit bar systems induced the creation of protected marshland areas and the formation of dune ridges systems on top of the spit bars. The marshland areas are the end product of the sedimentary infilling of the original Holocene basin/paleoestuary by the Guadalquivir river and other channel systems, a complex process that started to develop in the manner of a bird foot delta type formation in a low-energy environment (Rodríguez-Ramírez et al., 1996; Rodríguez-Ramírez, 1998). These marshes are fed by a main tributary (Guadalquivir River) and numerous ebb-tide channels or “caños” (Travieso, Guadiamar, Madre de la Marisma), with recent and former banks denoted by clay-rich levees as well as bioclastic and beach-morphology ridges (Carrizosa-Vetalarena, Las Nuevas, Vetalengua) (Fig. 1a). With a low gradient, features in the marshland include various landforms (levees, channels, point-bars) that have resulted from intense fluvial action and extensive sandy and shelly ridges (cheniers) resting on the clayey sediments, which have been shaped by marine processes, occasionally very violent, operating against the spit bars (Rodríguez-Ramírez et al., 1996). Prevailing winds are from the SW (Fig. 1b) and although winds from the SE and E are less common, they are important since they are commonly associated with storm events (Rodríguez-Ramírez, 1998). The sheet of aeolian sediments is composed of several generations of dunes (Fig. 1a) formed by westerly winds; some stabilized dune systems date to the Late Pleistocene (31–11.5 kyr cal BP, systems I and II), and others to the early to mid-Holocene (11–5/4 kyr cal BP, system III), while active dune systems (IV and V) have formed during the late Holocene (Rodríguez-Ramírez et al., 1996, 2005).

This study focuses on the marshland area (“marisma”), which includes the great alluvial plain and the ecosystem of greater extension of the National Park, covering an area of approximately 34,000 ha. This area, which has an average elevation of 1.35 m above sea level (asl), is formed by impermeable silt-clay soils (Fig. 1a). The climate of DNP is Mediterranean with an Atlantic influence, usually classified as dry subhumid, with wet, mild winters and dry, hot summers; the annual average rainfall is 580 mm, with high inter-annual fluctuations (Siljeström et al., 1994; Serrano et al., 2006). Although some areas are influenced by Atlantic tides in and out of the Guadalquivir River estuary, most of the marshland is free from tidal influence (Rodríguez-Ramírez et al., 1996). The marshland receives surface waters from the Guadiamar River basin and the Guadalquivir River estuary, but also from artificial water sources; the water entering these rivers and streams comes from the underlying aquifer as well as rainfall (Serrano et al., 2006). The hydrological pattern of the marshland exhibits a typical Mediterranean climate dynamic, with sudden and irregular flooding followed by long periods of confinement and dryness (Geertz-Hansen et al., 2010). The marshland follows a marked annual cycle, filling up with water in October, reaching an average depth of 20 cm, and remaining flooded until March/April, forming an inundated surface area. Afterwards, net water losses through evapotranspiration gradually dry out the marsh. By the end of the summer, only the deepest depressions retain highly saline waters, such as shallow ponds, gullies, and some channel stretches (Geertz-Hansen et al., 2010).

2.2. Modern vegetation

DNP comprehends three main ecosystems (Fig. 1b): the stabilized sands, the mobile dunes, and the salt marshes (Clemente et al., 1997;

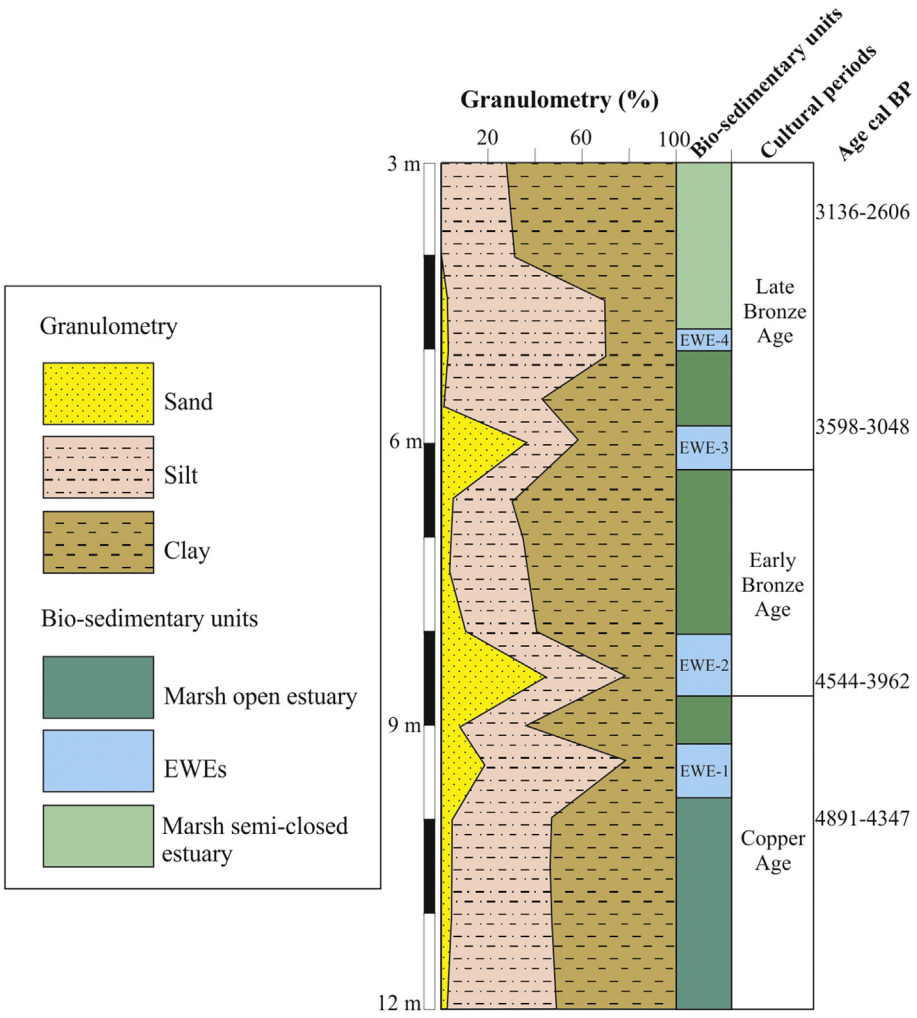

Fig. 2. Granulometry, bio-sedimentary units, calibrated ages (cal BP), and cultural periods of the S1 core.

García-Novo and Marín-Cabrera, 2006). The stable dune systems, which occupy almost a third of the total surface in El Abalario dune field (Fig. 1a), comprise ancient dunes that extend inland towards the NW limit of the marshland (Rodríguez-Ramírez et al., 1996). Their vegetation, locally known as "cotos" consists of extensive woodlands (Rivas-Martínez et al., 1980; Siljeström and Clemente, 1990). *Juniperus phoenicea* subsp. *turbinata*, *J. oxycedrus* subsp. *macrocarpa*, and *Pistacia lentiscus* characterize the driest and xerophytic enclaves, accompanied by the so-called "monte blanco" (scrubland made of *Cistus salvifolius*, *C. libatonis*, *Osyris quadripartita*, *Rosmarinus officinalis*, *Stauracanthus genistoides*, *Thymus mastichina*, *Lavandula stoechas*, *Helichrysum angustifolium*, *Halimium commutatum*, and *H. halimifolium*), while cork oak (*Quercus suber*), stone pine (*Pinus pinea*), *Phillyrea angustifolia*, and *Olea europaea* var. *sylvestris* thrive in moister areas, accompanied by the so-called "monte negro" (scrubland made of *Rubus ulmifolius*, *Ulex minor*, *U. australis*, and *Calluna vulgaris*, which, in particularly humid areas, incorporate hygrophytic species such as *Erica scoparia*, *E. ciliaris*, *E. umbellata*, *Saccharum ravennae*, and *Imperata cylindrica*). The most hygrophytic plants of the cork oak woodland include *Arbutus unedo*, *Pyrus bourgaeana*, *Myrtus communis*, *Smilax aspera*, *Rubia peregrina*, and *Lonicera periclymenum*. Ripparian communities consist mainly of *Tamarix africana*, *T. canariensis*, *Populus alba*, *Fraxinus angustifolia*, *Salix alba*, and *Ulmus minor*. Vegetation composition of the mobile dunes is dependent on the speed of (dune) movement and the depth of the water table, there being large areas of bare sand (Finlayson et al., 2008). The main woody species in the interdunal valleys are *Pinus pinea* and *Corema album*, while the dune crests are dominated by psammophilous communities of *Ammophila arenaria*, *Artemisia crithmifolia*, and *Armeria pungens*, as well as by scarce *Juniperus phoenicea* subsp. *turbinata* trees (Rivas-Martínez et al., 1980; López-Albacete, 2009).

The vegetation of the marshland (Fig. 1b) is determined by both the aforementioned seasonality and the microtopography (Rivas-Martínez et al., 1980; García-Viñas et al., 2005; López-Albacete, 2009). The low-lying zones that remain flooded for longer periods in the yearly cycle (6–8 months) are called "lucios", which constitute, with the partly-blocked fluvio-tidal channels ("caños"), the low marsh or "marisma baja" (Rodríguez-Ramírez, 1998). Helophyte swamp communities (freshwater marshes), dominated by species of Cyperaceae (*Scirpus maritimus*, *S. lacustris*, *S. littoralis*, *Eleocharis multicaulis*) and *Isoetes* spp., characterize the "caños" and their side areas ("quebradas"), while the "lucios" host macrophyte communities of both submerged - (*Callitriche* spp., *Myriophyllum alterniflorum*, *Ruppia drepanensis*) and floating aquatic macrophytes (*Potamogeton* spp., *Hydrocharis morsus-ranae*, *Ranunculus* subgen. *Batrachium*, *Lemna* spp.) (Díaz-Paniagua et al., 2010). Halo-nitrophilous communities (salt marshes), dominated by species of Chenopodiaceae (*Arthrocnemum macrostachyum*, *Salicornia ramosissima*, *Suaeda vera*), the so-called high marsh ("marisma alta"), characterize the highest elevation enclaves (1–3 m asl), known as "paciles" and "veras". "Paciles" are upper areas that have the appearance of emerged islands during heavy floods; "veras", which present a ground water seepage along the ecotone between the marshland and the "caños", are small raised areas of sandy soil that are relicts of old beaches or margins of the estuary and provide humidity to meadows and hydrophytic vegetation. This saline marsh vegetation dominates much of DNP due to

the enormous extension of the saline soils (García-Viñas et al., 2005; García-Novo and Marín-Cabrera, 2006), not only in the higher areas but also during the periods in which the marshland is not flooded (April–October). *Arthrocnemum macrostachyum* is the most abundant species in the DNP marshes, thanks to its ability to take advantage of subsurface hypersaline waters during the summer, or dry season, and its ability to resist shallow flooding during the rainy season (García-Murillo et al., 2007). In the months of rain and flooding, this vegetation is gradually replaced by freshwater marsh and aquatic communities in the lower areas, because of the dilution of surface salinity and the underground rhizomes that resist the dry season (Espinar, 2004).

3. Materials and methods

3.1. Geomorphology

Aerial photographs from 1956 (1:33,000) and 2013 (1:10,000) were analyzed to map the detailed geomorphology of landforms and sediments. The analysis was complemented by direct observations in the field. The Topographic Map of Andalusia (1:10,000) was used as a base document for geomorphological mapping and all information was integrated and analyzed within a project GIS (gvSIG).

3.2. Coring, sampling, sedimentological analysis and radiocarbon dating

A 1200 cm long core (S1, 36° 57′ 36.62″ N; 6° 23′ 8.80″ W, 1.5 m asl) was retrieved from the central part of the Doñana marshland (Fig. 1) in July 2016, using a rotation-drilling machine with continuous core sampling. Here we only present data for the lower 900 cm of the core (Fig. 2). The sediment core sections were placed in plastic tubes, protected in plastic guttering, and stored under cold conditions (4 °C) prior to laboratory sub-sampling and analysis. The stratigraphy and lithology of the core was described in the field. Grain-size distribution was determined in the laboratory by wet sieving for the coarser fractions (> 100 μm small) and by photosedimentation on a Mastersizer 2000 laser diffractometer ("Sedigraph 5100") for fractions smaller than 100 μm. The core was sectioned into continuous 10 cm-thick slices for pollen analysis.

Five samples of mollusk shell were radiocarbon-dated using the AMS technique. The dating was conducted by Beta Analytic Inc. (Miami, USA) and the Accium BioSciences AMS Laboratory (Seattle, USA). The marine shells selected were those that showed a low degree of transport or no transportation and had preserved and articulated valves. The dates were calibrated using CALIB 7.1 with the Marine13 curve (Reimer et al., 2013) (Table 1). Correction of the reservoir effect followed the specific guidelines suggested by Soares and Martins (2010) regarding calibration of marine shells from the Gulf of Cadiz; they have recommended a ΔR value of 100 ± 100 ^{14}C yr for the age range 4400–4000 ^{14}C yr BP. Because a lack of sufficient data for the next two millennia preclude determining with enough confidence the most recent time boundary to which the 100 ± 100 ^{14}C yr ΔR value can be extended, we chose to extend it to the middle year 3000 ^{14}C yr BP. New data on reservoir effects are necessary to calibrate with greater precision samples from the Gulf of Cadiz (Lario et al., 2011). An age-depth model was produced by using Clam 2.2 software (Blaauw, 2010); the best fit was obtained after applying a smoothing spline to the available radiocarbon dates. Confidence intervals of the calibrations and the age-depth model were calculated at 95% (2σ) with 1000 iterations. According to such a model, the base of the 1200 cm core can be assigned to the Copper Age (~5000 cal BP) and the sequence extends up to the present day. Terms and reference dates for classification into cultural periods are those of García Sanjuán and Odriozola (2012), who on the basis of accumulated ^{14}C datings for Southwest Iberia have proposed to distinguish between "The Copper Age" (to ~4200 cal BP), "The Early Bronze Age" (~4200–3500 cal BP), and "The Late Bronze Age" (~3500–2800 cal BP).

3.3. Pollen analysis

Pollen analysis was carried out on 90 sub-samples of 1 cm^3 along the lower 900 cm of the core. The core had a particularly good resolution for the Copper and Bronze Age, ~5000 cal BP to ~2800 cal BP, providing nearly decadal resolution (2.4 yr/cm). All samples were treated chemically (HCl, KOH, HF) following the standard procedures described by Moore et al. (1991), using Thoulet solution for densimetric extraction of pollen and non-pollen microfossils (Goeury and de Beaulieu, 1979). Acetolysis, however, was not applied, in order to allow identification of any contamination by modern pollen. Palynomorphs were identified at 400× and 1000× magnification by the use of the reference collection of the Institute of History at CSIC-Madrid, as well as identification keys and photo-atlases (Sáenz, 1982; Moore et al., 1991; Reille, 1999; van Geel, 2001; Beug, 2004; Cugny et al., 2010). Cistaceae pollen types were discriminated according to Sáenz (1979); *Pinus pinaster* and *Quercus suber* pollen differentiation followed Carrión et al. (2000a, 2000b). Pollen concentration (grains cm^{-3}) was estimated by adding a *Lycopodium* tablet to each sample (Stockmarr, 1971) and these values were divided by deposition time (year cm^{-1}) to calculate pollen accumulation rate (grains cm^{-2} year^{-1}). Pollen counts of up to 400 grains total land pollen (tlp) per sample were identified and counted. Pollen of aquatic or wetland plants as well as spores and non-pollen palynomorphs (NPPs) were excluded from the pollen sum. The pollen diagrams have been plotted against age, using TGview (Grimm, 2004).

3.4. Charcoal analysis

Charcoal debris were counted under the microscope alongside the identification of pollen and classified into two size classes (Vannière et al., 2008), namely > 125 and < 125 μm, interpreted as indicating local and regional fires, respectively (Whitlock and Larsen, 2001). Charcoal accumulation rates (CHAR) were calculated by sedimentation rates (cm year^{-1}) and expressed in particles cm^{-2} year^{-1} (Long and Whitlock, 2002). It is noted that pre-treatment processes can damage or split original large charcoal debris into smaller pieces. Yet, since all samples were processed using the same procedure, we assume that this systematic bias was uniform across samples and therefore did not influence the indicated temporal variation of charcoal abundance. Previous studies comparing the sieving charcoal series versus the pollen-slide charcoal series show that both of them display a similar pattern (Carcaillet et al., 2001; Yin et al., 2016).

Table 1
AMS-radiocarbon data with 2σ range of calibration from S1 core.

Laboratory code	Depth (cm)	^{14}C age BP	Age cal BP	Median	ΔR (^{14}C yr)
D-AMS 006934	200	3053 ± 22	2966–2416	2721	100 ± 100
D-AMS 006935	360	3153 ± 29	3136–2606	2842	100 ± 100
Beta-288897	580	3560 ± 40	3598–3048	3331	100 ± 100
D-AMS 006936	850	4286 ± 30	4544–3962	4266	100 ± 100
Beta-288898	1000	4560 ± 40	4891–4347	4636	100 ± 100

3.5. Paleontology

Macrofossil analysis from sediment samples was undertaken in order to identify mollusk species type and diversity. Furthermore, mollusk shell taphonomy was assessed. Thirty-three samples were collected from the core and prepared by washing the bulk sediment (12 cm^3) through a 1 mm sieve. Bivalves and gastropods were identified to species level and then counted to study the semi-quantitative distribution of species along the studied core interval. The presence and relative abundance of other groups (such as scaphopods, barnacles, and bryozoans) were also noted.

4. Results

4.1. Facies identification, core lithology and chronostratigraphy

The lowest part of core S1 (Fig. 2), from 1200 to 970 cm (~5000–4560 cal BP), suggests a typical marsh estuary sequence of mostly grey-greenish (10YR6/1) clayey silts (52–55% clay, 42–44% silts, and 2–5% sand). The mollusk assemblage appears to be dominated by shallow-water estuarine bivalve species from sheltered environments, including *Tellina* spp. and *Crassostrea angulata*; *Cerastoderma edule* – typical from lagoonal environments – and *Barnea candida* appeared in a minor proportion. The gastropod class is characterized by greater species diversity, though with a very low density of specimens. The species found are *Nassarius reticulatus*, *Rissoa* spp., *Bittium reticulatum*, *Cerithium* spp. and *Bulla* spp. In general, the mollusk macrofauna indicates minimal transportation, particularly bivalves; both valves were present, some of them even in life position (Ruiz et al., 2005b; Rodríguez-Ramírez et al., 2015). Open marine species – *Venus* spp. – were very sparse and found scattered; they were probably transported to the study area. Relatively low sediment accumulation rates characterize this earliest part of the S1 record, with values of around 5.22 mm/yr.

In the S1 record we were able to identify a first EWE at a depth of 970–930 cm (EWE-1: ~4560–4450 cal BP) from the significant amount of sand (15–20% sand). This first sand layer (Fig. 2) consists of grey (10YR6/4) clayey silty sands (55–60% silts, 22–30% clay). The macrofauna is characterized by a mixture of disarticulated valves, shell fragments, and whole bivalves, of a high species diversity characteristic of marine environments as well as those that are typical of the inner estuary. The bivalvia class is represented by a greater abundance of *Cerastoderma edule*, *Tellina tenuis*, *Tellina* spp., *Crassostrea angulata*, *Neopycnodonte* spp., *Ostrea edulis*, *Barnea candida*, *Spisula solida*, *Solen* spp., *Venus* spp., *Chlamys multistriata*, *Nucula nucleus*, *Pecten* spp., *Donax* spp., *Chamelea gallina*, *Anomia ephippium*, *Loripes lucinalis*, and *Tapes decussatus*. Regarding the gastropod class, *Nassarius reticulatus*, *Rissoa* spp., *Murex brandaris*, and *Bittium reticulatum* stand out, while *Calyptraea chinensis* appears sporadically. Some specimens of scaphopods, bryozoans, and crayfish remains are also present. Further up, between 930 and 860 cm, the amount of sand in the core progressively decreases (10%), while the amount of clay increases and silt decreases within grey-greenish (10YR6/1) clayey silts (Fig. 2). In this facies ~4450–4200 cal BP the macrofauna is exactly the same as the one described for the first facies of the record, ~5000–4560 cal BP, suggesting again a closed paleoestuary with hardly any marine influence. Low sediment accumulation rates (2.80 mm/yr) also characterize this part of the S1 record. At the top of this facies, between 900 and 860 cm (Fig. 2), sand levels reach a minimum (7%), as do silts (28%), while clays increase considerably (65%), though the faunal composition remains the same as in the previous facies.

A second EWE is record in the lithostratigraphy ~4200–4000 cal BP – between 860 and 800 cm (EWE-2; Fig. 2); the layer comprises grey (10YR6/4) heterometric sands (45% sand content) within a matrix containing a relatively high percentage of sandy silts (33%) and clay (22%). Low sediment accumulation rates (3.00 mm/yr) characterize this part of the S1 record. The macrofauna includes the same abundance of mollusk species described for the first EWE event ~4560–4450 cal BP.

Later, ~4000–3500 cal BP (800–615 cm), the granulometry and the macrofauna are again the same prior to EWE-2, showing a matrix with high percentages of clay (70%) and, to a lesser extent, silts (25%), including 5% of sands (Fig. 2), and low sediment accumulation rates (3.70 mm/yr).

Between ~3500 and 3435 cal BP, a new EWE (EWE-3) is documented in the core by a sand layer within grey (10YR6/4) clayey silts, between 615 and 590 cm (35% sand, 42% clay, 23% silts). The layer exhibits a fining-upward succession of sediments, its boundary with the lower formation of silty clays being slightly erosive (Fig. 2). A sediment accumulation rate of 3.80 mm/yr characterizes this part of the S1 record. The macrofauna is similar to that of the two previous sandy facies, mentioned above: the layer shows an increase in articulated and disarticulated bivalve shells and shell fragments within a high diversity of characteristic species from marine environments as well as others typical of the inner estuary. Later, ~3435–3185 cal BP – between 590 and 500 cm – the sedimentary facies (Fig. 2) is characterized by a typical marsh estuary sequence of mostly grey-greenish (10YR6/1) clayey silts (43–74% silts, 30–57% clay, 1–2% sand), while the mollusk macrofauna appears to be dominated by the shallow brackish estuarine species described above, with relatively low sediment accumulation rates around 3.60 mm/yr.

A fourth EWE (EWE-4) is documented in the S1 record by a small accumulation of sands (5%) within the clayey silts (65% silts, 30% clay) between 500 and 480 cm (Fig. 2), that is, ~3185–3140 cal BP. The sediment accumulation rate increased to 4.44 mm/yr. This facies includes a muddy-sandy layer overlying an erosive base that contains an accumulation of shells and shell fragments, mostly coming from marine mollusk species (although there are also estuarine species). Further up core S1 (480–300 cm; ~3140–2800 cal BP), the sedimentation changes progressively into grey-ochre silty clays (10YR8/4) (27–67% silts, 30–73% clay, 0–3% sand), while the accumulation rate increases to 5.29 mm/yr (Fig. 2). This part of the core exhibits a scant macrofauna in terms of both number of individuals and number of species: within the bivalvia class, *Scrobicularia plana*, *Cerastoderma edule*, *Tellina tenuis*, and *Tellina* spp.; within the gastropod class, only *Rissoa* spp.

4.2. Pollen, non-pollen palynomorphs and charcoal

The results of pollen, non-pollen palynomorphs (NPPs) and charcoal analyses are displayed in Figs. 3–5. To establish the zonation of the pollen sequence, we tested several divisive and agglomerative methods with the programme IBM SPSS Statistics 21. Based on the ecological meaning of the obtained zones, nine local pollen assemblage zones (LPAZ-1 to LPAZ-9) were constructed on the basis of agglomerative constrained cluster analysis of incremental sum of squares (Coniss) with square root transformed percentage data (Grimm, 1987). The number of statistically significant zones was determined by using the broken-stick model (Bennett, 1996). A total of 109 pollen, spore, and non-pollen palynomorph types were identified. The results of the identification and counting are presented as percentage diagrams (Figs. 3–4). Fig. 5 shows both microscopic and macroscopic CHARs related to selected pollen and NPP percentage curves. The terms 'local' (0–20 m), 'extra-local' (20 m–2 km), and 'regional' (> 2 km) used in the text refer to different pollen source areas according to Prentice (1985). The pollen proportion (Fig. 3) of *Pinus pinaster* (< 3%), *Pinus sylvestris/nigra* (1–4%), and deciduous *Quercus* (< 7%) can be attributed to long distance ('regional') transport from mountainous areas (López-Sáez et al., 2010a, 2010b, 2013, 2015). However, it should not be ruled out that some dispersed specimens of *Pinus pinaster* or *P. nigra* could be from trees that lived in the DNP area during the Holocene, since abundant macrofossils of both species have been found in Upper Pleistocene aeolian deposits exposed in the Asperillo cliff (Postigo-Mijarra et al.,

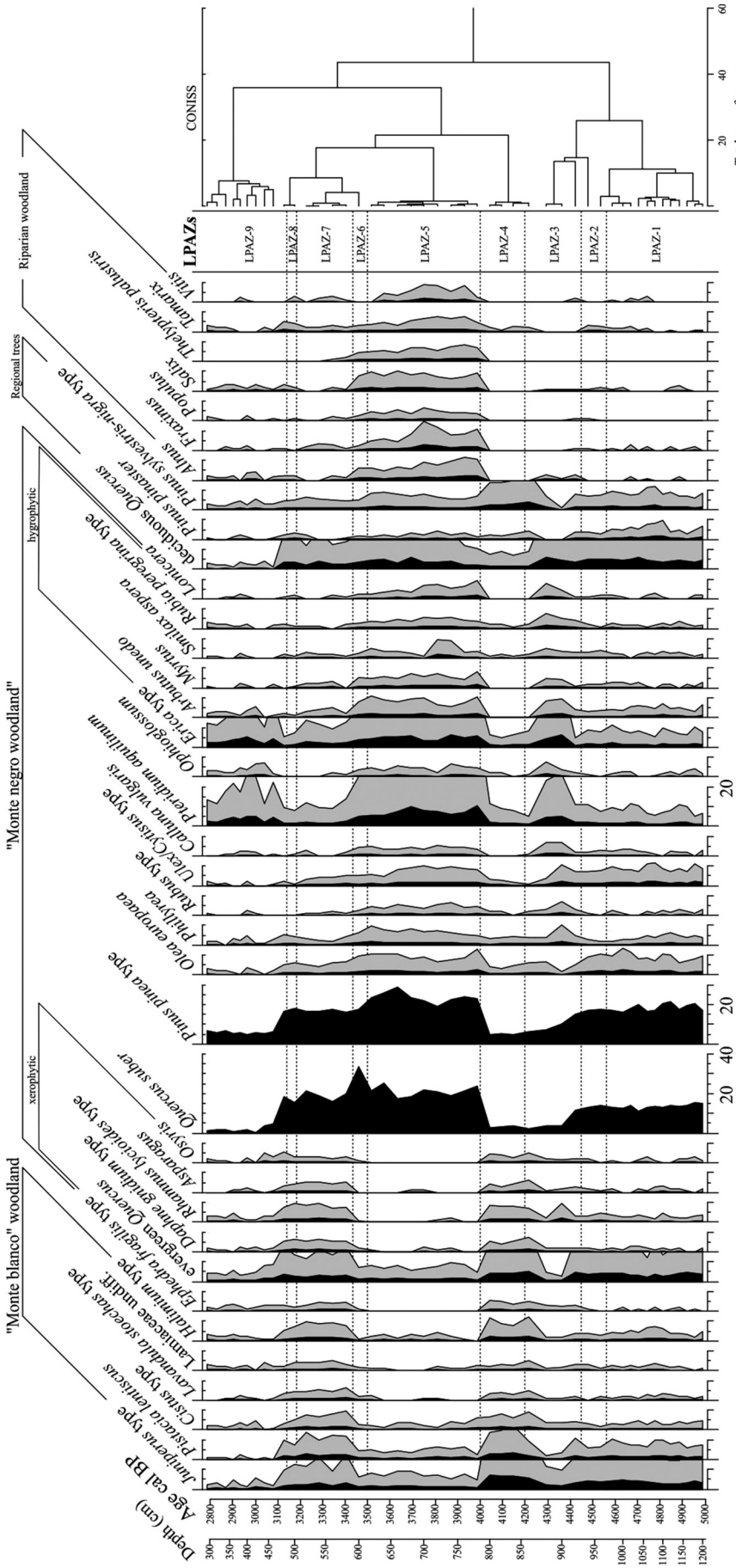

Fig. 3. Percentage pollen diagram of the S1 core (woodland vegetation) plotted against age (cal BP). The black silhouettes show the percentage curves of the taxa, the grey silhouettes show the ×5 exaggeration curves. Groups of pollen were established according to their ecological affinities, following Galiano and Cabezudo (1976) and Rivas-Martínez et al. (1980).

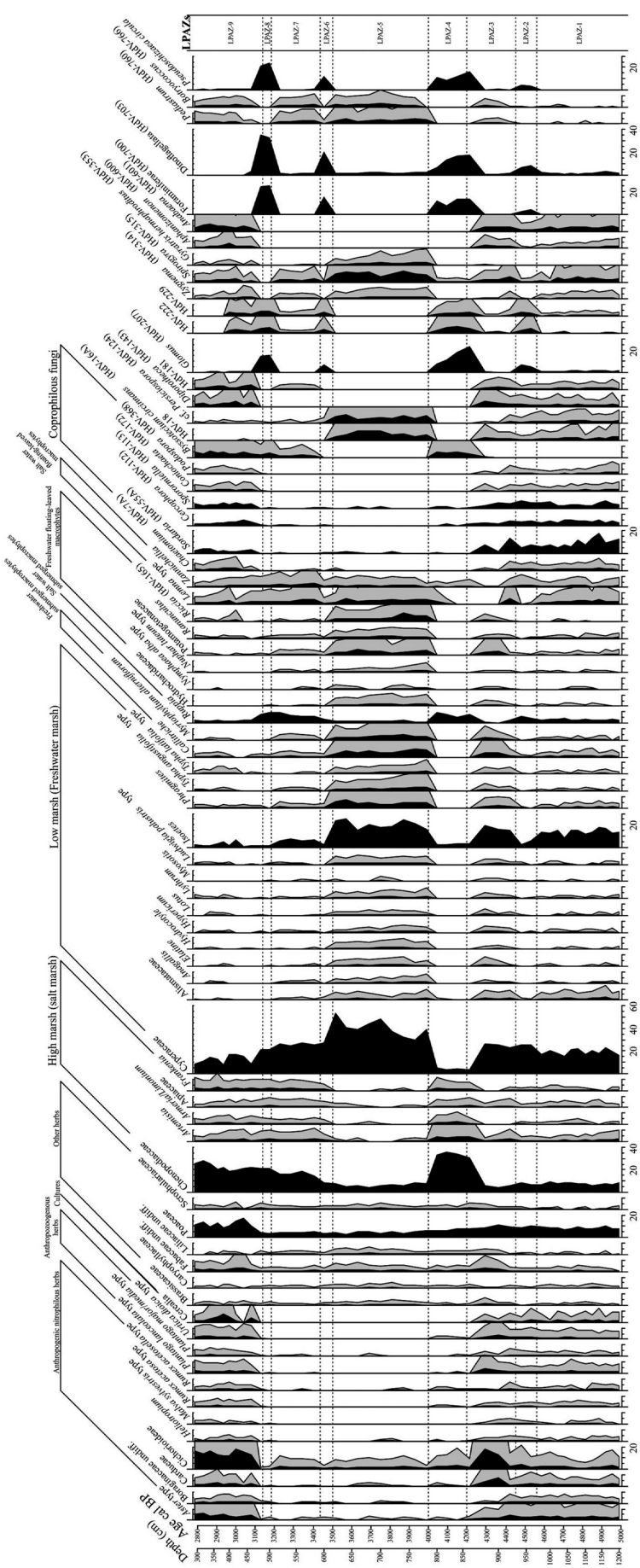

Fig. 4. Percentage pollen diagram of the S1 core (herbs, hydro-hygrophytic taxa, ferns and non-pollen palynomorphs) plotted against age (cal BP). The black silhouettes show the percentage curves of the taxa, the grey silhouettes show the ×5 exaggeration curves. Groups of pollen and non-pollen palynomorphs were established according to their ecological affinities, following Galiano and Cabezudo (1976), Rivas-Martínez et al. (1980), van Geel (2001) and López-Albacete (2009).

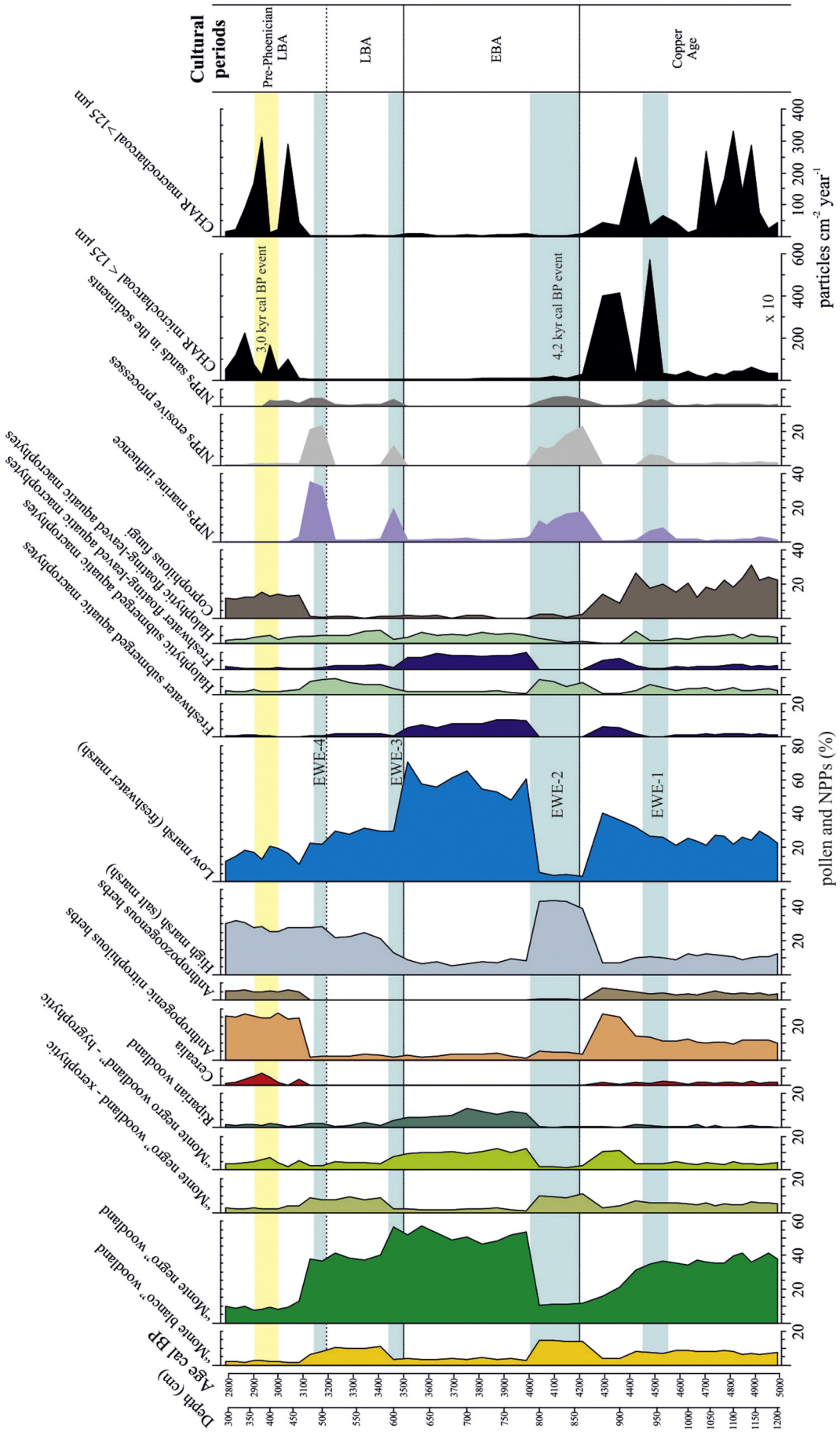

Fig. 5. Cultural periods, EWEs, climatic events, percentage pollen diagram (selected pollen and NPP taxa), and charcoal accumulation rates (CHARs) plotted against age (cal BP) from the S1 core.

2010; Morales-Molino et al., 2011), as have been wood remains from *Pinus pinaster* on Matalascañas beach that date from the late Holocene (Rodríguez-Ramírez et al., 2012).

Geobotanical interpretation of stone pine woodlands (*Pinus pinea*) in the DNP have led to controversial interpretations between those who accept the natural character of these woodlands (Costa-Tenorio et al., 1997) and those who reject it (Rivas-Martínez et al., 1980; Asensi and Díez-Garretas, 1987). This debate is still ongoing, despite a growing body of evidence from historical and palaeoecological studies that point to the native origin of these regional stone pine forests (Martínez and Montero, 2004). In Southwest Iberia, macrofossil and palynological records show the presence of stone pine for at least the last 50 kyr (Stevenson, 1984, 1985; López-Sáez et al., 2001, 2002a; Carrión et al., 2008), suggesting the permanence of widespread coastal pine woodlands since the Upper Pleistocene. *Pinus pinea* type is present in the S1 pollen record from the mid-Holocene ~5000 cal BP (Fig. 3), demonstrating the natural character of stone pine woodlands in DNP; this is also in accordance with natural habitat predictions for the mid-Holocene derived from machine learning models (Benito-Garzón et al., 2007).

In reference to the grapevine (*Vitis vinifera*), its presence is more or less constant throughout the S1 pollen record (Fig. 3), although always sporadically and in low percentages (0–2%). Similar percentages of grapevine pollen have been documented in the mid-Holocene record from El Acebrón (Fig. 6) (Stevenson and Moore, 1988; Stevenson and Harrison, 1992), which the authors interpret as the existence of wild populations along stream corridors; however, the recurrence of pyrophilous pollen taxa such as *Cistus ladanifer* leads them to consider the possibility of local cultivation. This latter hypothesis is also supported by Stevenson (1985) from the documentation of exceptionally high percentages of *Vitis* (> 40%) in the pollen record from Las Madres (Fig. 6), which led this author to argue for the local cultivation of grapevine in the Doñana area between the Late Neolithic and the Copper Age. However, a later study of this same sequence points out the possible misidentification between *Vitis* and *Rhamnus* pollen types (Yll et al., 2003). In short, should we assume the existence of wild populations of grapevine (*Vitis vinifera* subsp. *sylvestris*) in the DNP (Cantos et al., 2017); the data provided by the S1 pollen record (Fig. 3) probably indicate the natural existence of the species in riparian forests with *Salix atrocinerea* and *Thelypteris palustris* (Rivas-Martínez et al., 1980).

5. Discussion

5.1. Copper Age (~5000–4200 cal BP)

The S1 core sediment record started ~5000 years ago (Table 1). The geomorphological scenario ~5000–4200 cal BP (Rodríguez-Ramírez et al., 1996, 2014, 2015; Ruiz et al., 2005b) is a more or less closed paleoestuary with little marine influence and permanent fresh water input, partially protected along its westernmost boundary by aeolian units and to the south and southeast by the growing Doñana barrier spit (Fig. 6c). By ~4200 cal BP, the extension of the paleoestuary may have been less than half of what it was at the time of the Postglacial Maximum, ~6500 cal BP, making it a brackish lagoon in practice. North of this lagoon, the original paleoestuary had turned into marshland, the oldest marshland formed in the lower Guadalquivir river valley during the Holocene (Fig. 6c).

The aforementioned data agree with the pollen spectra of LPAZ-1–5000–4560 cal BP (Figs. 4–5), where the low marsh dominates locally (21.1–29.7%). The dominant vegetation was Cyperaceae (15.6–22.6%) grasslands, with species able to withstand certain seasonal changes in salinity and relatively brackish waters, such as *Scirpus maritimus*, *S. littoralis*, and *Alisma plantago-aquatica* (Alismataceae, 1.1–2.6%) among the helophytes, which would thrive in silty clay soils with some sand content (Rivas-Martínez et al., 1980). The continued presence of cf. *Persiciospora* (HdV-124) is related to helophyte marsh with eu- to mesotrophic conditions (Pals et al., 1980). Aquatic submerged macrophytes such as *Ruppia* (2.5–4.1%) and *Zannichellia* developed in brackish areas of deeper water, while on the surface there were dense floating aquatic macrophytes lawns (*Lemna* 1.6–4.5%) capable of living in relatively saline waters. High marsh plants (9.1–12.7%) would populate the edges of the lagoon, being composed mainly by Chenopodiaceae, *Artemisia*, *Armeria/Limonium*, and *Frankenia*. In addition, in lower brackish areas, or in freshwater pools fed by fluvial inputs, communities of submerged macrophytes (~2%) dominated by *Callitriche* and *Myriophyllum alterniflorum*, accompanied by other pollen taxa of floating macrophytes (2–3%; Hydrocharitaceae, *Nymphaea alba*, *Nuphar luteum*, Potamogetonaceae, *Ranunculus*, *Riccia*) developed. High values of *Isoetes* (8–18%) – a fern that lives in seasonally flooded soils – are noteworthy in this pollen zone, pointing out the prevalence of shallow waterlogged environments in the proximity of the core site (Valdés et al., 2007). The constant presence, though in low values, of dinoflagellata cysts (< 3%) can be explained by a certain marine influence inside the paleoestuary. In short, ~5000–4560 cal BP the marsh would comprise a mosaic of diverse plant communities, determined by salinity and microtopography.

From a palynological point of view, the lithostratigraphic and macrofaunal record of EWE-1 ~4560–4450 cal BP corresponds to pollen zone LPAZ-2 (Fig. 4), which is characterized by the appearance of Foraminiferae (2.6–4%), the increase of Dinoflagellata (7–8%), Cyperaceae (25%), and *Ruppia* (4–6%) and a significant drop (< 1%) of both freshwater floating and submerged macrophytes (Fig. 5). High values of *Glomus* (5–7%) and *Pseudoschizaea circula* (4–5%) can be correlated with erosive processes on land derived from EWE-1 (Ruiz et al., 2005a), whereas percentage increases (~4%) of both HdV-222 and HdV-229 are related to the contribution of sand in the sedimentary deposit (van Geel et al., 1989; López-Sáez et al., 1998). All of these data suggest the existence of a paleoestuary still confined ~4560–4450 cal BP, but with greater marine influence. It is the first time that an EWE has been detected in this chronological range in the DNP, although the record could also be probably related to the storm surge with spit barrier breaching described by Lario et al. (1995, 2010, 2011) and Rodríguez-Ramírez et al. (2015) for ~4500–4200 cal BP in the Guadalquivir marshland. Probably, EWE-1 would have had no more than a local effect – the sediment accumulation rate (3.63 mm/yr) being even lower than in the preceding phase – and, therefore, would not have affected the prehistoric settlement of the marshlands. In fact, the S1 pollen record (Figs. 4–5) shows the continuous presence of cereal pollen with high values (1–2.5%) as well as notable pastoral pressure (coprophilous fungi 18–20%) ~4560–4450 cal BP; furthermore, the relative stability of both “monte negro” and “monte blanco” woodland pollen assemblages ~5000–4200 cal BP is not disrupted ~4560–4450 cal BP (Fig. 5).

Between ~4450 and 4200 cal BP (LPAZ-3) the vegetation is very similar to that described in LPAZ-1 (Figs. 3–4). Cyperaceae (27%) and *Isoetes* (19%), as well as many other typical low marsh or freshwater pollen taxa (Alismataceae, *Elatine*, *Hypericum*, *Lotus*, *Lythrum*, *Ludwigia palustris*, *Phragmites*, *Typha angustifolia*, and *T. latifolia*), increase within helophytes, while *Callitriche* (3.7%) and *Myriophyllum alterniflorum* (2.5%) also increase within freshwater-submerged macrophytes at the top of LPAZ-3, ~4360–4200 cal BP. Freshwater floating macrophyte taxa are also well represented, particularly Potamogetonaceae (3%), while *Lemna* disappears and *Ruppia* slightly decreases (Figs. 4–5). These data can be related to a humid phase described for the western Mediterranean ~4350–4150 cal BP, during which rainfall increased considerably (Magny, 2004; Magny et al., 2006; López-Sáez et al., 2017). In the case of the DNP, such rainfall increase would mean a greater contribution of fresh water to the marshland, increasing the low marsh (40%) and decreasing the high marsh (7%), producing also an increase of algal populations (*Botryococcus*, *Pediastrum*, Zygnemataceae) and other NPPs (HdV-18, HdV-181, *Gyratrix hermaphroditus*); these taxa are signs of meso-eutrophic moist environments (López-Sáez et al., 1998;

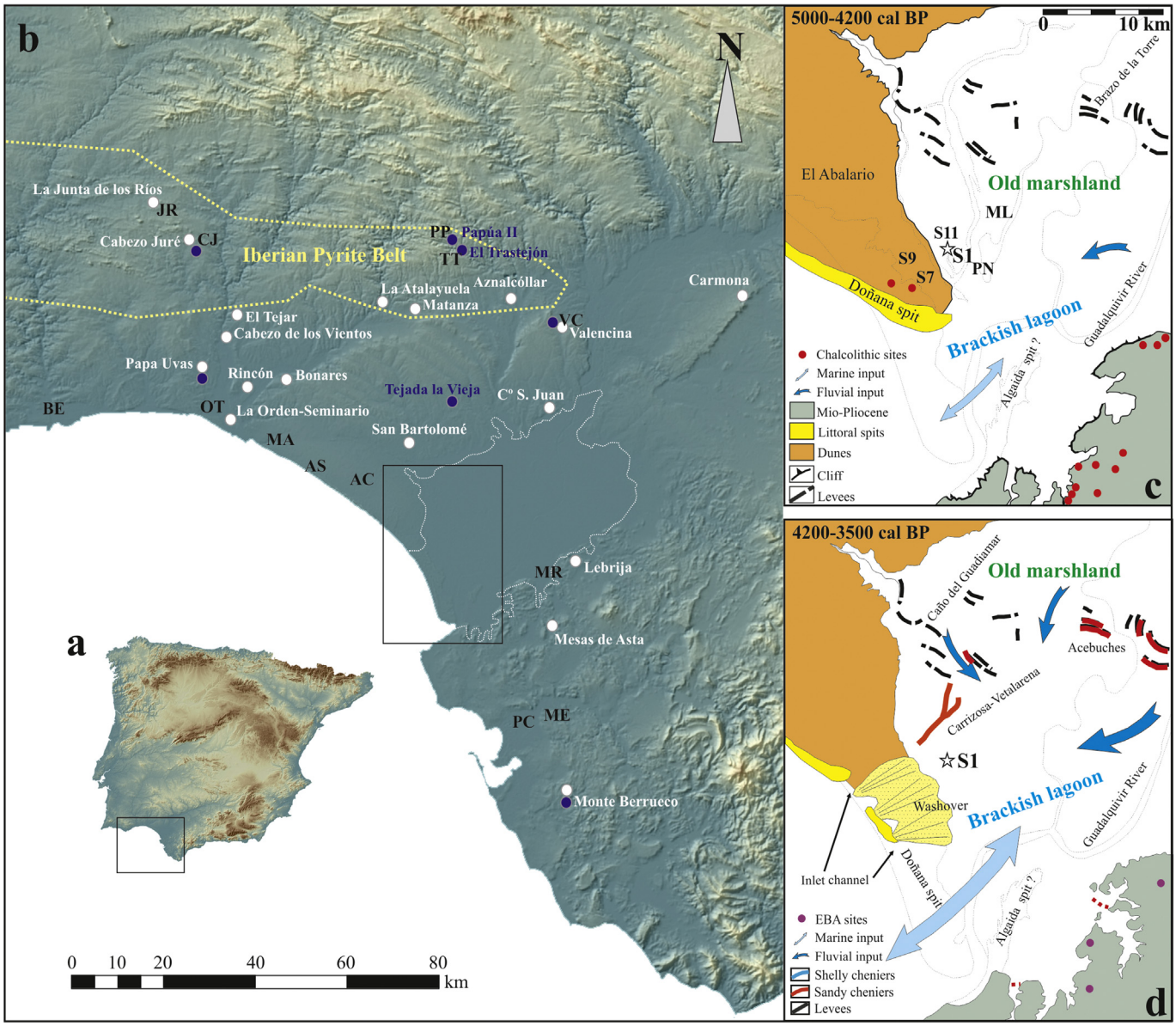

Fig. 6. (a) Geographic framework of the study area: southwestern Iberian Peninsula. (b) Main Chalcolithic (white circles) and Early Bronze Age archaeological sites (blue circles) on the southwestern Iberian corner. Dotted white line represents the hypothetical position of the Guadalquivir paleoestuary (Arteaga et al., 2016) and the black rectangle the study area (Doñana National Park). Location of the S1 core (white star) and geomorphological scenario of the study area (c) ~5000–4200 and (d) ~4200–3500 cal BP, respectively, following Rodríguez-Ramírez et al. (2014, 2015). Palaoenvironmental records mentioned in the text: AC (El Acebrón), AS (Asperillo), BE (Beliche), CJ (Cabezo Juré), JR (La Junta de los Ríos), MA (Las Madres), ME (Medina Lake), ML (Marilópez), MR (Marismillas), OT (Odiel-Tinto estuary core), PC (Pocito Chico), PN (PN core), PP (Papúa II), S7 (S7core), S9 (S9 core), S11 (S11 core), TT (El Trastejón), VC (Valencina de la Concepción). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

van Geel and Aptroot, 2006). This humid phase is also well represented in the S1 pollen record by the development of the more hygrophytic "monte negro" facies (Fig. 5).

During the mid-Holocene (~5000–4200 cal BP; LPAZ-1 to LPAZ-3), extra-local DNP forest landscapes, according to data from S1 record, are characterized by the presence of poorly-developed "monte negro" woodlands, with *Quercus suber* (4–15%), *Olea europaea*, and *Pinus pinea* (7–22%) as the main taxa, probably occupying the stable dunes of El Abalario that are closest to the lagoon (Fig. 6c). Shrubs were abundant, with *Calluna vulgaris*, *Arbutus unedo*, *Erica*, *Ulex/Cytisus*, *Myrtus*, *Rubus*, *Smilax aspera*, *Rubia peregrina*, *Lonicera*, and *Phillyrea* being the most important taxa from "monte negro" shrubland, and *Cistus*, *Ephedra fragilis*, *Daphne gnidium*, *Rhamnus lycioides*, *Asparagus*, *Osyris*, *Juniperus*, *Pistacia lentiscus*, *Halimium*, and Lamiaceae from "monte blanco" shrubland (Figs. 3 and 5). *Pteridium aquilinum* (1–5%) and *Ophioglossum* are two ferns that are also characteristic of "monte negro" shrubland (Fig. 3). *Quercus suber* percentages are fairly low ~5000–4200 cal BP (Fig. 3), which indicate a "dehesa-type" landscape (López-Sáez et al., 2010a). LPAZ-1 to LPAZ-3 are also characterized by significant frequencies of anthropogenic nitrophilous herbs (*Aster*, Boraginaceae undiff., Cardueae, Cichorioideae, *Heliotropium*, *Malva sylvestris*, *Rumex acetosa*, *R. acetosella*) and anthropozoogenous types like *Plantago lanceolata* (1–4%), *P. major/media* (~1%), and *Urtica dioica* (1.5–4%), as well as by the continuous presence of coprophilous fungi such as *Chaetomium*, *Sordaria*, *Cercophora*, *Sporormiella*, *Coniochaeta*, and *Podospora* (Figs. 4–5); all of which suggests seasonal grazing activities and significant human pressures (Galiano and Cabezudo, 1976; Cabezudo, 1979; Behre, 1981; López-Sáez and López-Merino, 2007), associated with a large settlement ~5000–4200 cal BP at the DNP (Fig. 6c).

Cerealia (0.5–2.6%) show a continuous record ~5000–4200 cal BP

(Figs. 4–5), revealing extra-local agriculture (López-Sáez and López-Merino, 2005). On the eastern paleocliffs and plateaux bordering the coastal lagoon (Fig. 6c) where the soils are more fertile than in the El Albalario dune zone, to the west (Bejarano, 1997; García-Rivero, 2004), numerous Chalcolithic sites have been identified (Arteaga and Roos, 1992). However, considering the geographical position of S1 (Fig. 6c) and the relatively high values (> 2%) of cereal pollen ~5000–4950, 4885, 4815, 4670, and 4535 cal BP (Fig. 4), it is likely that agriculture was practiced also in the sandy western area of the Doñana marshland during this time, especially in those areas of the floodplain where the concentration of surface salt was low. In parallel, the continuous presence ~5000–4200 cal BP of Cyanobacteria *Anabaena* (2–4%) and *Aphanizomenon* (0.5–2.5%) and that of ascospores of *Diporotheca* (2–5%) (Fig. 4) indicates major soil disturbance, extensive soil erosion, and phosphate-eutrophication due to the impact of agricultural activities, and livestock trampling of marshland ecosystems (López-Sáez et al., 1998; van Geel et al., 1994, 1996; Hillbrand et al., 2012).

Some authors contend that these Chalcolithic communities of the lower Guadalquivir basin lived mostly on fish and shellfish, supposedly placing agricultural plots only on the eastern and northern edges of the Guadalquivir paleoestuary (Escacena, 2001, 2014). However, regional paleodietary studies drawing on stable isotope analyses reveal a homogeneous diet based on C3 terrestrial plant and animal resources – meat, milk, and dairy products combined with pulses and cereals – with minor ingestion of aquatic foodstuff (Fontanalls-Coll et al., 2015). The data provided by the S1 pollen record are conclusive and in line with the paleodietary evidence: during the Chalcolithic, a mixed agricultural and livestock economy was practiced in the DNP area, at least on both the eastern and western boundaries of the marshland, during a long chronological interval of almost a millennium (~5000–4200 cal BP). Surely, these crops must have been grown in those areas with fertile soils (along the eastern edge) or in those places where fresh water was available, yet also in some enclaves of this type existing in the ecosystem mosaic of the DNP marshes, as evidenced by both freshwater-submerged and floating aquatic macrophytes in the S1 pollen record (Figs. 4–5). Perhaps such crops were grown as well on sandy extensions of the El Abalario field or on an early stage of the Doñana spit, today deeply buried (Fig. 6c).

The “dehesa” system – open savannah-like woodlands – represents a cultural landscape shaped by combined human and livestock pressure, i.e. an anthropozoogenous landscape resulting from farming and grazing activities in the oak forests (*Quercus ilex* and *Q. suber*). This system of agricultural exploitation of the forest originated in the western half of Iberia between the Late Neolithic and the Chalcolithic (Stevenson and Harrison, 1992; López-Sáez et al., 2005, 2007), linked to new concepts of landscape organization represented by the first megalithic tombs (García Sanjuán, 1999, 2005; Díaz-Zorita et al., 2012). The existence of a “dehesa” landscape in the DNP marshland during the Copper Age (~5000–4200 cal BP) was the outcome of a rational, long-term management of the agroforestry resources, entailing a lasting sedenterisation of the population. In fact, areas with greater agricultural capacities, such as wetlands, were increasingly populated in southwestern Iberia from the Early Chalcolithic (~5200 cal BP) (Balsera et al., 2015; Lillios et al., 2016), with very dense settlement of farmsteads, preferably in the valley floors and in the fertile countryside (García-Rivero, 2004), as cereal farming spread (Acosta, 1995; Mederos, 1996).

From ~4500 cal BP onwards, settlements diversified (Fig. 6b): some sites are small (1–5 ha) and are located on hills or in low-lying settings, comprising both open and walled sites (Cabezo de los Vientos, La Orden-Seminario, El Tejar, Rincón, and Papa Uvas in the Odiel-Tinto paleoestuary; Cerro de San Juan, Carmona, San Bartolomé de Almonte, Bonares, Mesas de Asta, and Lebrija in the vicinity of the Gualquivir paleoestuary; Monte Berrueco in the Guadalete estuary); other sites are large and unenclosed (> 50 ha) or represent so-called ditched megasites such as Valencina de la Concepción, across around 450 ha (Escacena and De Frutos, 1986; García Sanjuán and Hurtado, 1997; García-Sanz and Fernández-Jurado, 1999; Belén et al., 2000; Pérez-Macías et al., 2002; Abril, 2003; García-Rivero and Escacena, 2015; Garrido and Vera, 2015; García Sanjuán et al., 2017). At this time numerous Copper Age settlements stood at the mouth of the Guadalquivir river on the eastern edge of the DNP marshes (Arteaga and Roos, 1992, 1995; Fig. 6c). Abundant archaeological remains from the Late Neolithic and the Chalcolithic have also been found on the western active dune system of the DNP (Campos et al., 1993; Borja et al., 1999). The increase in both the total number of sites and the average size of the sites suggests that the Chalcolithic expansion was accompanied by a substantial growth of the population (Blanco-González et al., 2018). In fact, this period was the climax of a rising demographic trend favoured by a surplus economy practiced during the Late Neolithic (Acosta, 1995), driven in turn by technological advances such as the plough and animal traction (Sherrat, 1981; Harrison, 1985; García Sanjuán and Hurtado, 1997). In addition, zooarchaeological data from sites such as Papa Uvas and Valencina de la Concepción indicate the importance of bovines, pigs, and ovicaprines at that time (Harrison and Moreno, 1985; Martín de la Cruz, 1994), which agrees with the aforementioned strong pastoral pressure detected in the S1 pollen record.

Human and livestock impact on coastal woodland areas as well as expansion of agriculture have also been tracked ~5000–4200 cal BP in pollen records from nearby sites in Southwest Iberia (Fig. 6b–c), such as El Acebrón, Las Madres, Asperillo, Marilópez, Marismillas, and S11 in the DNP and its vicinity (Caratini and Viguier, 1973; Stevenson, 1984, 1985; Stevenson and Moore, 1988; Stevenson and Harrison, 1992; Zazo et al., 1996; Yll et al., 2003; López-Sáez et al., 2011; Jiménez-Moreno et al., 2015), as well as at the archaeological site of Pocito Chico in the Cadiz Bay (López-Sáez et al., 2001, 2002a, 2002b). In the case of the El Acebrón peat bog pollen record (Stevenson and Moore, 1988), the above-mentioned human imprint during the 5th millennium cal BP correlates with sedimentary bands rich in macrocharcoal, which would indicate the use of fire for deforestation by human communities. These data coincide with high concentrations of CHAR macrocharcoal particles ~4890–4780 cal BP (143–331 particles $cm^{-2} yr^{-1}$) and ~4700 cal BP (269 particles $cm^{-2} yr^{-1}$) in the S1 record (Fig. 5).

The study area is only 40 km away from the Iberian Pyrite Belt (Fig. 6), one of the richest metallogenic zones of Western Europe (Mellado et al., 2006). Local metallurgy dates back to the first centuries of the 5th millennium cal BP (Leblanc et al., 2000; Abril, 2003; Nocete et al., 2005, 2011); however, its scale and environmental impact are subject to ongoing debate (e.g., García-Alix et al., 2013). Some authors have argued for woodland-consuming copper production activities on a relatively large scale in the Aracena mountain range (Nocete et al., 2005, 2011); they claim that copper metallurgy conducted at Chalcolithic sites such as Cabezo Juré or Valencina de la Concepción would have caused a significant environmental impact. Deforestation of the landscape and pollution by heavy metals at both local and regional levels were the ultimate consequences of intensive metal mining activities (Nocete et al., 2005, 2008; Nocete, 2014). While diverse paleoecological records might support this view, their interpretation is not straightforward and other, simpler interpretations of the evidence are possible. These other scholars argue that copper metallurgy at this time was a localised domestic activity of part-time crafts people (Hunt, 2003; Costa et al., 2010; Murillo-Barroso et al., 2015). Although pollen analyses from Cabezo Juré, La Junta de los Ríos, and Valencina de la Concepción (Fuentes et al., 2006; Nocete et al., 2004–2005, 2008) (Fig. 6b) show a certain increase in deforestation – the presence of poorly-developed oak woodlands – and erosive processes, metallurgy might not have been the only contributing activity. Holocene sediments from the Odiel-Tinto estuary contain high metal (Cu, As, Pb, Au, Hg) concentrations in layers dated to 4480 cal BP (Leblanc et al., 2000; Nocete et al., 2005). Moreover, in a study of the sedimentary infill of the Guadiana estuary Delgado et al. (2012) determined that anthropogenic sources of metals (indicated by enrichment of Pb, Co, Ni, Mn,

and Cu) prevailed over natural sources at that time (~4500 cal BP). Nevertheless, more recent literature (García-Alix et al., 2013) challenges the interpretation of such anomalies in cultural terms. In the DNP, the geochemical analysis of the PN core (Carretero et al., 2010, 2011), very close to our S1 record (Fig. 6c), documented the first evidence of slight palaeocontamination ~4500 cal BP, probably related to the presence of settlements keen on working the ore deposits of the Iberian Pyrite Belt near the Agrio River in the vicinity of Aznalcóllar (Fig. 6b), a tributary of the Guadiamar River (Hunt, 2003; Gómez-Toscano, 2006; Tornos, 2008). The magnitude and traceability of such a pollution episode, however, must have been restricted to a local scale, since such observations have no supporting evidence in marine records in the Alboran Sea or in further terrestrial cores (Martín-Puertas et al., 2010; García-Alix et al., 2013). On the other hand, the S1 record registers increase in CHAR microcharcoal particles from ~4500 cal BP onwards, with three maximum values (569, 413, and 399 × 10 particles cm^{-2} $year^{-1}$, respectively) ~4480, 4360, and 4290 cal BP (Fig. 5). These values may be due to fire-related activities in the Aracena range from ~4500 cal BP, including forest clearance and herding in addition to metallurgy. In short, pre-Bronze Age signatures of heavy metal pollution and deforestation remain open to a variety of conflicting explanations (García-Alix et al., 2013).

5.2. Early Bronze Age (~4200–3500 cal BP)

The Copper Age–Early Bronze Age transition ~4200–4000 cal BP was a threshold period in the landscape history of the DNP marshes. In the S1 record, the pollen zone LPAZ-4 records a sharp decline of low marsh (3–5%) and *Isoetes* (2.3–3.7%) percentages and the disappearance of both freshwater floating- and submerged-aquatic macrophyte pollen taxa (Figs. 4–5); this suggests the advent of a markedly arid climate phase, with salt concentration at the ground surface. This phase is characterized by the spread of halophytic herbaceous (high marsh) pollen taxa such as Chenopodiaceae (30–35%), *Artemisia* (3.6–4.5%), Apiaceae, *Armeria/Limonium*, and *Frankenia*, and the spread of submerged-aquatic macrophytes typical of saline or brackish waters such as *Ruppia* (5–9%), *Lemna* (2.3%), and *Zannichellia* (Fig. 4). On a regional scale, the decrease in deciduous *Quercus* percentages (< 2%) and the parallel increase in the evergreen *Quercus* values (4–5%) are also documented (Fig. 3). Locally, in this period the "monte blanco" pollen taxa increase their values (14–15%) – particularly *Juniperus*, *Pistacia lentiscus*, *Cistus*, and *Halimium* –, probably due to their xerophytic character (Figs. 3 and 5), while those of "monte negro" decrease significantly (*Quercus suber* 2.4–3.7%, *Pinus pinea* 5–6.3%). The joint occurrence (Fig. 4), in low percentages, of *Spirogyra* and *Byssotecium circinans* (3–5%) may well be indicative of meso- to euthropic conditions in the marshland during a dry phase (van Geel et al., 1989; López-Sáez et al., 1998; van Geel and Aptroot, 2006).

During the transition from the 5th to the 4th millennia cal BP the study area was subjected to the effects of a short, abrupt climate change of extreme aridity, known as the "4.2 kyr cal BP event" (Magny, 1993, 2004) or "Bond Event 3" (Bond et al., 2001), which has been recognized as a marked aridification phase in the Mediterranean Iberian region (Jalut et al., 2009; Carrión et al., 2010). Evidence of the 4.2 kyr cal BP event (~4200–4000 cal BP) is present in the pollen record of the S11 core from the DNP (Jiménez-Moreno et al., 2015), at El Acebrón and Las Madres in the Doñana territory (Stevenson and Moore, 1988; Stevenson and Harrison, 1992; Yll et al., 2003), at Pocito Chico in the Cadiz Bay (López-Sáez et al., 2001, 2002a, 2002b), and in the Beliche marshland in the Guadiana estuary in southern Portugal (Fletcher et al., 2007) (Fig. 6b–c); this evidence consists of increases in Chenopodiaceae and xerophytic taxa such as *Juniperus* and *Artemisia*, and conversely decreasing values of wetland taxa (Cyperaceae, *Isoetes*, *Myriophyllum*). A paleoclimatic reconstruction from the neighboring S11 pollen record indicates an abrupt decrease in the average annual rainfall of at least 45 mm, which in turn, coincides with maxima in temperature parameters ~4100 cal BP (Lillios et al., 2016). A similar process happened in Medina Lake, as indicated by a marked lowering of the lake level and a loss of the diatom record ~4100 cal BP (Reed et al., 2001).

Nevertheless, ~4200–3800 cal BP (likely average date: ~4000 cal BP) a large extreme wave event (EWE) has also been documented, identified as a tsunami in other parts of Southwest Iberia. In the DNP marshland it is recorded as an interbedded marine layer made up of fine storm deposits and thicker tsunami-induced shelly deposits; it is documented within the inner marsh deposits of the Guadalquivir paleoestuary (Rodríguez-Ramírez et al., 1996; Rodríguez-Ramírez and Yáñez, 2008; Lario et al., 2010, 2011). The occurrence of this EWE has also been inferred from the lithostratigraphy of the S1 core ~4200–4000 cal BP (EWE-2; Fig. 2) as well as from the neighboring borings of S7, S9, and S11 cores (Fig. 6c); it is characterized by sandy layers that contain massive accumulations of shells (both articulated and disarticulated bivalves) and shell fragments in a sandy-muddy matrix with gravel and lithoclasts, on an erosive base (Rodríguez-Ramírez et al., 2015). Very high values of *Glomus* (8–23%) and *Pseudoschizaea circula* (9–16%) can be also correlated with erosive processes derived from EWE-2 (Fig. 4), whereas percentage increases of HdV-222 (3–6%) and HdV-229 (1.7–3.3%) are related to the sandy deposits (van Geel et al., 1989).

The temporal coincidence of a climatic change with a high energy marine incursion of the possible tsunami in the DNP ~4200–4000 cal BP (Fig. 5) is likely to have had an effect on the Chalcolithic communities that occupied the territory. EWE-2 might have destroyed some of the settlements and reworked their remains across the estuarine basin (Rodríguez-Ramírez et al., 2015). This hypothesis is plausible if one takes into account that EWE-2 caused major geomorphological changes that are symptomatic of tsunamigenic processes of a large magnitude; these signs include (Fig. 6d) the breaching and sudden erosion of the Doñana spit barrier – including a large overwash of the southernmost section of the neighboring aeolian system of El Abalario – and the generation of large sandy cheniers (Carrizosa-Vetalarena, Los Acebuches) (Rodríguez-Ramírez et al., 2015; Celestino-Pérez et al., 2016). The rather catastrophic effects in the Guadalquivir estuary were compounded by the intense neotectonic subsidence detected in the area (Rodríguez-Ramírez et al., 2014).

The consequences of both climate change and the tsunamigenic event ~4200–4000 cal BP are clearly reflected in both the palynological and the archaeological records of the transitional period between the end of the Chalcolithic and the onset of the Early Bronze Age in the DNP. From a palynological point of view, in addition to what has already been commented on, there are almost no traces of anthropogenic impact on vegetation in the S1 sequence, neither in the charcoal nor in the pollen record (Figs. 3–5). Coprophilous fungi show a strong downward trend (0.4–2.6%), especially *Sordaria*, *Sporormiella*, and *Cercophora*, while *Chaetomium* and *Podospora* disappear. In fact, the sporadic presence and low percentages of coprophilous fungi must be interpreted as a result of the movement of wild animals through the marshland and not domesticated livestock. Anthropogenic nitrophilous (3–5%) and anthropozoogenous pollen taxa (< 1%) sharply decrease, suggesting a very low or no human impact and the end of arable (absence of cereal pollen) and pastoral activities. Furthermore, high values of Dinoflagellata (6–18%) and Foraminiferae (2.24%) suggest oceanic movements driven by high energy, which would have allowed these planktonic organisms to reach the interior of the paleoestuary. A similar picture can be gathered from other pollen records in Southwest Iberia (Fig. 6b–c), such as S7, S11, El Acebrón, Las Madres, Pocito Chico, Medina, Beliche, Quarteira, Carcaxai, Alcantarilha, and Farelo-Torre (Stevenson and Moore, 1988; Stevenson and Harrison, 1992; López-Sáez et al., 2001, 2002a, 2002b; Reed et al., 2001; Yll et al., 2003; Fletcher et al., 2007; Jiménez-Moreno et al., 2015; Schneider et al., 2016). From an archaeological point of view, the final result was the collapse of the Chalcolithic agrarian landscape in the DNP, a major decline in the number of settlements in coastal areas, and the

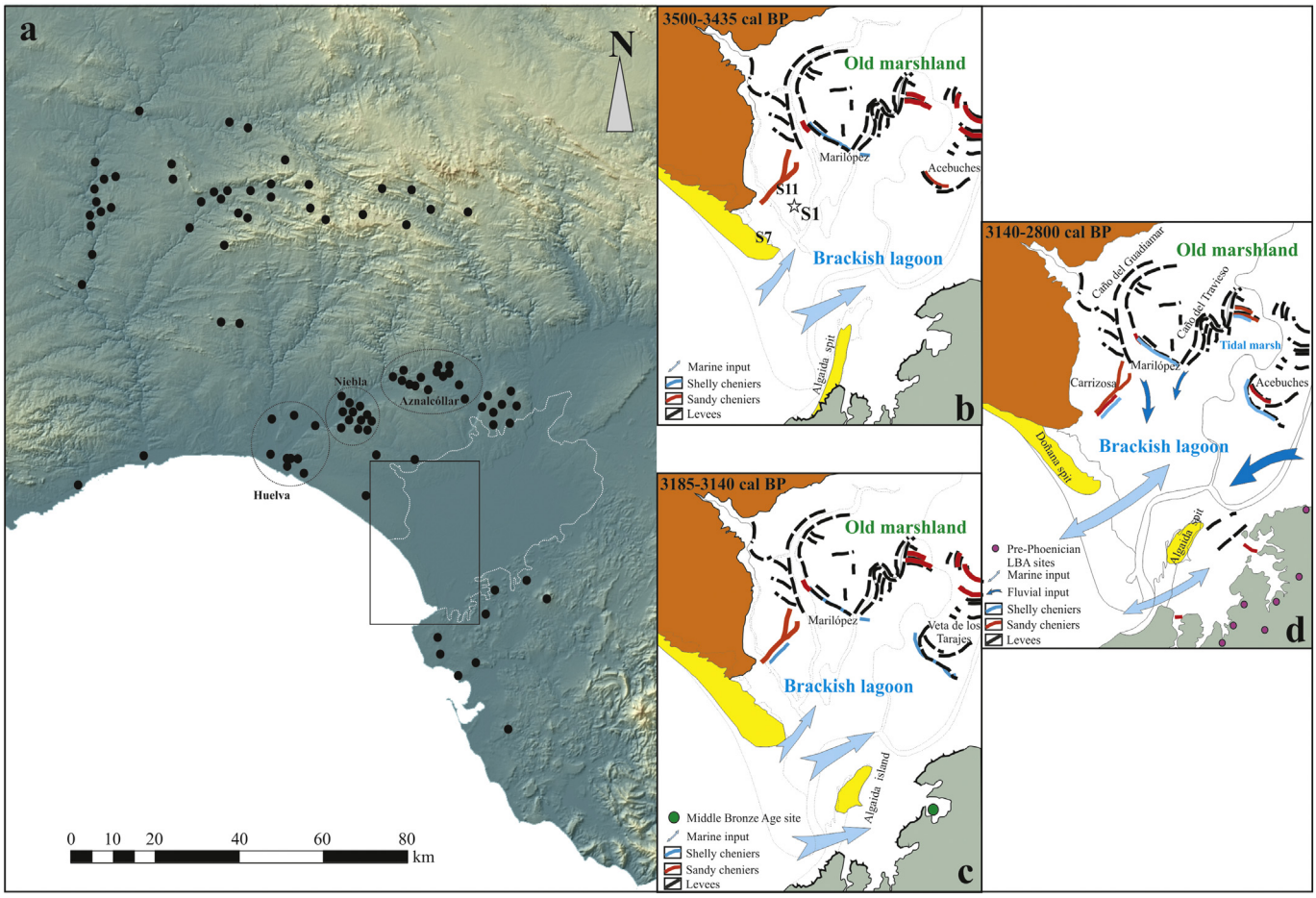

Fig. 7. (a) Nuclear areas of pre-Phoenician Late Bronze Age (~3200–2800 cal BP) archaeological sites in the southwest of the Iberian Peninsula (modified from Gómez-Toscano, 2008). Dotted white line represents the hypothetical position of the Guadalquivir paleoestuary (Arteaga et al., 2016) and the black rectangle the study area (Doñana National Park). Location of the S1 core (white star) and geomorphological scenario of the study area (b) ~3500–3435, (c) ~3185–3140 and (d) ~3140–2800 cal BP, respectively, following Rodríguez-Ramírez et al. (2014, 2015). Palaoenvironmental records mentioned in the text: S7 (S7 core), S11 (S11 core).

emergence of a more pastoralist-oriented economy in mountain environments (e.g. the Aracena range) during the Early Bronze Age (Lillios et al., 2016; Blanco-González et al., 2018).

Subsequently, ~4000–3500 cal BP (LPAZ-5, 800–615 cm), the marshland ecosystem recovered to the vegetation conditions observed prior to EWE-2 (Figs. 3–5). While low marsh (48–70%) and *Isoetes* (15–26%) notably increased, high marsh taxa sharply decreased (< 10%). In addition, both freshwater-submerged (7–10%) and floating (6–10%) aquatic macrophytes reappeared, as did other freshwater hydrophytic elements such as *Phragmites* (3.5–7%), *Typha angustifolia*, and *T. latifolia*. Within NPPs, *Pediastrum* (2–4.3%), *Botryococcus* (2–3%), *Zygnema* (1–2%), *Spirogyra* (5–10%), *Gyratrix hermaphroditus* (2–3%), HdV-124, and HdV-18 reappeared (Fig. 4), suggesting humid and mesotrophic conditions (van Geel et al., 1989; van Geel and Aptroot, 2006). These data can be correlated with the humid phase described by Magny (1993, 2004) in the western Mediterranean ~4000–3500 cal BP. This stage would correspond to a period of relative stability of the marsh ecosystems, with higher rainfall and low sediment accumulation rates. Although the DNP paleoestuary may still have included a brackish lagoon, a certain saline influence is perceived in the record by the constant presence in it of Dinoflagellata in low percentages (Fig. 4). Pollen evidence of anthropization is still absent ~4000–3500 cal BP, which is consistent with very low percentages of both anthropogenic nitrophilous (< 4%) and anthropozoogenous herbs (0–0.5) and coprophilous fungi (< 2%).

At the onset of the Early Bronze Age (EBA), ~4200 cal BP, cultural changes occurred. Most Copper Age settlements were abandoned and new ones, such as El Trastejón and Papúa II (Fig. 6b), were established in the Aracena range ~4200–3500 cal BP (Hurtado et al., 2011), although such villages are poorly understood (García Sanjuán and Odriozola, 2012; Lillios et al., 2016). Moreover, there is a significant lack of EBA layers overlying the previous Chalcolithic occupations, suggesting the displacement of populations and their relocation elsewhere (García-Rivero and Escacena, 2015). Unlike the Chalcolithic settlement pattern, during the EBA the habitats are smaller and appear scattered throughout the study area (Fig. 6b), occupying very heterogeneous ecosystems between the coast (Monte Berrueco), the countryside (Tejada la Vieja, Papa Uvas, Valencina de la Concepción, Setefilla), and the mountains (Cabezo Juré) (Aubet et al., 1983; Escacena and De Frutos, 1985, 1986; Calderón et al., 1987; Caro-Bellido, 1988; Martín de la Cruz, 1994; Mederos, 1996; Nocete et al., 2005; García-Rivero and Escacena, 2015). Such a new settlement pattern is particularly evident on the eastern edge of the Doñana marshes (Fig. 6c–d), where the number of EBA settlements is rather low when compared to those of the previous Chalcolithic period (Caro-Bellido, 1982, 1989; Arteaga and Roos, 1992, 1995).

These reasons, besides the ephemeral nature of habitats, have led some authors (García Sanjuán, 1999; Ruiz-Gil and López-Amador, 2001; Harding, 2003; López-Sáez et al., 2014) to posit that EBA communities had an economy with a spectrum of means of subsistence that was narrower than that of their Chalcolithic predecessors, who probably relied more on livestock than on arable agriculture and followed a more mobile lifestyle, with part of the population depending on herding strategies. Indeed, pollen records from El Trastejón and Papúa II

(Fig. 6b) show a landscape during the EBA that was little affected by human impact and basically shaped by pastoral activities (high values of anthropozoogenous pollen taxa), with insignificant evidence of cereal pollen (Martín-Consuegra and Ubera, 2011). From ~3700 cal BP onwards the availability of fertile zones near the EBA sites seems to be a location factor of minor importance, whereas their strategic position in hilly inexpugnable spurs gained importance (Campos and Gómez-Toscano, 1997; García Sanjuán, 1999). New prominent settlements were established in the EBA, some with massive terraces and stone-walls. The best-known site is El Trastejón, which features masonry slopes and terraces, with oval huts yielding evidence of copper smelting (Hurtado et al., 2011). In short, the Copper Age-EBA transition in the DNP is marked by an important decline of a human footprint. It is tempting to propose that the marsh environment suffered a substantial decline in population at the beginning of the Bronze Age ~4200 cal BP and those communities with different cultural backgrounds relocated to territories further inland. This suggested movement of local people is in line with the so-called 'replacement model' which envisages a cultural disruption at the Chalcolithic-EBA transition (García-Rivero and Escacena, 2015) and has led some authors to reconsider migration from the southwest to the southeast of the Iberian Peninsula after ~4500 cal BP as a plausible contributing factor to macro-regional dynamics (Lillios et al., 2016).

5.3. Late Bronze Age (~3500–2800 cal BP)

Between ~3500 and 3435 cal BP, a third EWE (EWE-3) is documented in the S1 record. This event has also been documented (Fig. 7b) in the neighboring records from S7 and S11 ~3550 cal BP (Rodríguez-Ramírez et al., 2015). The sedimentological and faunal features of this third EWE make it difficult to establish whether it was a tsunami or a storm surge, however. Because of its central position in the paleoestuary, like S11, point S1 was more exposed to marine influence than S7, which was protected by the Doñana spit (Jiménez-Moreno et al., 2015). Northeast of S1, the Marilópez chenier system contains shells and shell fragments as a result of estuarine resedimentation associated with storms, tsunamis, currents, and other processes (Rodríguez-Ramírez et al., 1996; Ruiz et al., 2005b; Rodríguez-Ramírez and Yáñez, 2008). Other sandy littoral strands, such as those of Veta de Los Acebuches (Fig. 7b), also contain shells derived from different environments that have been dated to ~3882–3334 cal BP (Rodríguez-Ramírez and Yáñez, 2008), thus indicating an estuary unrestricted by sandy barriers up to at least ~3300 cal BP.

From a palynological point of view, EWE-3 is correlated with pollen zone LPAZ-6, which registers (Figs. 3–5) a percentage increase of NPPs that indicate erosive events (*Glomus*, *Pseudoschizaea circula*). The sand-rich deposits (HdV-222, HdV-229) are augmented in high marsh pollen taxa (13%) and *Ruppia* (3.8%), and there is a diminution of low marsh pollen taxa (29.3%), *Isoetes* (2.6%), and freshwater-submerged and floating aquatic macrophytes (< 1%). LPAZ-6 is also characterized by the reappearance of Foraminiferae (15.1%) and an increase of Dinoflagellata (19.6%), which suggests a paleoestuary relatively confined ~3500–3435 cal BP but with remarkable marine influence. The percentage of anthropogenic nitrophilous herbs (1.4%), anthropozoogenous herbs (0%), and coprophilous fungi (1.2%) is very low, indicating the absence of any type of human impact on the ecosystems of the marshland at this time.

In the post-EWE-3 phase, ~3435–3185 cal BP (LPAZ-7), the high marsh becomes more important (21–24.6%), while the low marsh (28–31.4%) and *Isoetes* (7–8%) seem to be reduced; qualitatively, however, the low marsh presents a high number of pollen taxa, like the freshwater-submerged (2%) and floating aquatic macrophyte pollen taxa (3%), which suggests the coexistence of very diverse communities in the marshland itself (Figs. 4–5). In fact, other aquatic macrophytes capable of withstanding a certain degree of salinity (*Ruppia* 6–9.4%, *Zannichellia* 2–3.6%, *Lemna* 2.4–4.3%) also thrived at this time. Furthermore, "monte blanco" woodlands increase their values (10–11%), to the detriment of "monte negro" ones, which, nonetheless, remain the dominant woodland types in the DNP (Fig. 5). As in the previous phase, the palynological evidence of anthropization is insignificant. The co-occurrence of *Byssotecium circinans* (5–11%), HdV-181 (4–5%), *Pediastrum* (9–13%), *Botryococcus* (8–11%), *Zygnema* (4–5%), and *Spirogyra* (9–13%) indicates meso- to euthropic conditions in the marshland during a marked arid phase in the brackish lagoon that occupied the paleoestuary (van Geel et al., 1989; López-Sáez et al., 1998; van Geel and Aptroot, 2006). The presence of sand in the sequence (Fig. 2) justifies the recurrence of HdV-222 and HdV-229 in the pollen record, while some marine influence is recorded by the presence, albeit in low percentages, of Dinoflagellata (4–7%). All of these data can be correlated with the generalized climate aridification that affected the western Mediterranean during the mid-late Holocene, which seems to be due to the decreasing trend in summer insolation, lower sea-surface temperatures, and lower precipitation during the autumn-winter season (Magny, 1993, 2004; Jiménez-Moreno et al., 2015).

Later, a new EWE (EWE-4) is documented in the S1 record ~3185–3140 cal BP, which has been also documented in the S7 and S11 records ~3150 cal BP (Rodríguez-Ramírez et al., 2015). By taking into account the date of this event and its stratigraphic position, the EWE-4 deposit at point S1 can be attributed to a single event that spread across the paleoestuary and adapted to its paleogeography (Fig. 7c). The pollen spectra related to this fourth event (LPAZ-8) are very similar to those of LPAZ-6 (Figs. 3–5). However, unlike the previous EWEs, this one does seem to have had an important effect on the coastal woodlands of the DNP, since LPAZ-8 registers a decrease of both "monte blanco" (6.6–8%) and "monte negro" pollen taxa (< 40%). Regression is more pronounced in salt-sensitive taxa (*Quercus suber*, evergreen *Quercus*) and seeders ("monte blanco" pollen taxa such as *Pistacia lentiscus*, *Cistus* or *Halimium*) (Figs. 3 and 5), due to seed bank burial or destruction (Miyamoto et al., 2004); by contrast, more salt-tolerant taxa (*Pinus pinea*, *Olea europaea*) and sprouters ("monte negro" pollen taxa such as *Rhamnus lycioides* or *Phillyrea*) seem to be less affected by the ingress of saline waters into the marshland. These facts corroborate the hypothesis that the marine flood could have varied impacts on the terrestrial vegetation of the DNP, where the individual response of each taxa would be related to ecological features such as salt tolerance, biotype, and interspecific competitiveness. In turn, the high marsh becomes more important (28–29%), while the low marsh and freshwater aquatic macrophytes are significantly reduced. The erosive potential of EWE-4 is confirmed by maximum percentages of *Glomus* (14–16%) and *Pseudoschizaea circula* (21–23%), while the marine influence that it caused is documented by the reappearance of Foraminiferae (24.6–25.4%) and a maximum of Dinoflagellata (32.4–35.3%) (Fig. 4). Furthermore, LPAZ-8 registers no evidence of anthropization.

Such morphostratigraphic and palynological features lead us to consider EWE-4 as tsunamigenic, especially considering the distance of the three cores (S1, S7, S11) to the paleocoastaline (10–15 km), as well as the fact that similar events have been documented in the Tinto-Odiel and Guadalete paleoestuaries (Morales et al., 2008, 2011; Lario et al., 2010, 2011). EWE-4 must have brought about large and catastrophic geomorphological changes throughout the southwest of the Iberian Peninsula. In the Guadalquivir paleoestuary (Rodríguez-Ramírez et al., 2014, 2015) it caused extensive erosion of the Doñana spit and a rupture of the Algaida spit which transformed this spit into an island (Fig. 7c). Indeed, a high-energy layer has been found by means of shallow drilling of sediments in the eastern sector of the Guadalquivir river bank, which appears to have been generated by this event (Rodríguez-Ramírez et al., 2015); in the marsh of Rajaldabas this layer contains remains – at almost 1 m depth – of an archaeological site dated, by artefact typology to the "Bronze II espagnol" ending abruptly ~3200 cal BP (Menanteau, 1981) (Fig. 7c). Such abrupt change most likely occurred in other settlements in the DNP and certainly at nearby

Lebrija, on the eastern paleocliffs of the paleoestuary (Tejera-Gaspar, 1985; Caro-Bellido et al., 1987). However, as previously noted, the pollen evidence of such other potential settlements in the DNP is practically nonexistent. The marsh of Rajaldabas is some 20 km away from the marsh of Hinojos and point S1.

Judging by the macrofaunal record in different cores (S1, S7, S11), the Guadalquivir paleoestuary became confined after EWE-4 (Rodríguez-Ramírez et al., 2015). Input of transported marine molluscs decreased and species from sheltered environments became dominant (Celestino-Pérez et al., 2016). Definitely, the transition from an open estuary to a semi-closed one can be dated in the S1 record to just after ~3185–3140 cal BP and could be related to coastal progradation and the formation of sandy barriers in the river mouth (Fig. 7d). These facts are clearly discernible in the S1 pollen record by the disappearance of Dinoflagellates and Foraminiferae from ~3080 cal BP onwards (Fig. 4).

After EWE-4, the low-energy marine dynamics at work in the Gulf of Cadiz resumed their action: the Doñana spit started to grow again, even more towards the southeast than before, and fluvial inputs within the paleoestuary were reactivated, with the consequent stability of the marshland ecosystems. In fact, ~3140–2800 cal BP (LPAZ-9) climatic conditions are those of a constant aridification process from ~3000 cal BP onwards, which means that high marsh developed (25–32%), while low marsh (10–20%), *Isoetes*, and aquatic freshwater macrophytes were significantly reduced (Figs. 4–5). In contrast, aquatic vegetation capable of withstanding a certain degree of salinity underwent a small increase or remained at values akin to the preceding pollen zone. The joint occurrence of *Spirogyra* (1–3%), *Zygnema* (0.5–2.5%), HdV-181 (1–4.3%), *Gyratrix hermaphroditus* (~1%), *Pediastrum* (1–2%), *Botryococcus* (1–2%), and *Byssotecium circinans* (3–6%) could indicate meso- to euthropic conditions in the marshland during a dry period (van Geel et al., 1989; López-Sáez et al., 1998; van Geel and Aptroot, 2006), while the presence of both HdV-222 and HdV-229 until ~2965 cal BP could relate to the existence of sands (Fig. 2) in the S1 lithostratigraphy (van Geel et al., 1989).

Unlike the previous pollen zone, in LPAZ-9 the evidence of anthropization is very compelling ~3140–2800 cal BP (Fig. 5): anthropogenic nitrophilous (24–28%) and anthropozoogenous herbs (5–6%) sharply increase, as do coprophilous fungi (11–15.4%); furthermore, cereal pollen reappears in the sequence, sometime – ~3080 and 2965–2865 cal BP – with values above 3%, which would imply admitting, again, the existence of agricultural crops in the immediate vicinity of the S1 core (López-Sáez and López-Merino, 2005). In parallel, both "monte blanco" (< 3%) and "monte negro" (*Quercus suber* < 2%, *Pinus pinea* 5–7%) seem to degrade, although certain pollen taxa characteristic of the degradative stages of these formations increase (Rivas-Martínez et al., 1980), such as *Erica* (2–5%) and *Pteridium aquilinum* (3–5.3%), as well as Poaceae (9–17%) (Figs. 3–4). All of these developments chronologically coincide with an important settlement increase in the DNP environment during the pre-Phoenician Late Bronze Age (Fig. 7d). The reappearance of *Anabaena* (3–7%), *Aphanizomenon* (1–4%), and *Diporotheca* (2–5%) ~3140–2800 cal BP (Fig. 4) indicates extensive soil erosion and phosphate-eutrophication due to the impact of palaeoeconomic activities by LBA people (López-Sáez et al., 1998; van Geel et al., 1994, 1996; Hillbrand et al., 2012). These facts coincide with high concentrations of CHAR macrocharcoal particles ~3040 cal BP (289 particles $cm^{-2} yr^{-1}$) and ~2935–2900 cal BP (167–313 particles $cm^{-2} yr^{-1}$) in the S1 record (Fig. 5), which would allow us to suggest the anthropogenic use of fire on a local and extra-local scale, both for growing crops and for clearing vegetation to create more pasture for cattle.

Our data from LPAZ-9 are supported by other pollen records from Southwest Iberia which show similar trends, such as those from El Acebrón, Las Madres, Marilópez, and S7 and S11 in the DNP and its vicinity (Stevenson, 1984, 1985; Stevenson and Moore, 1988; Stevenson and Harrison, 1992; Zazo et al., 1996; Yll et al., 2003; Jiménez-Moreno et al., 2015). They are also supported by the archaeological site of Pocito Chico in the Cadiz Bay, where the last centuries of the Bronze Age show a very evident human impact – high values of anthropogenic nitrophilous herbs and cereals, decrease of the arboreal cover – as a consequence of population increase (López-Sáez et al., 2001, 2002a, 2002b). In addition, the arid event ~3000 cal BP has been locally recognized by the desiccation of Medina Lake (Reed et al., 2001); globally, the period ~3140–3000 cal BP coincides with "Bond cold event 2" from the North Atlantic (Bond et al., 2001) and low lake levels in central Europe and the western Mediterranean (Magny, 1993, 2004).

From a material culture viewpoint, it is necessary to distinguish the earliest centuries of the Late Bronze Age (LBA) (~3500–3200 cal BP) from the latest, or Pre-Phoenician, LBA (~3200–2800 cal BP). Only a few radiocarbon dates have been obtained in the region for this long period (García Sanjuán and Odriozola, 2012), to be supplemented by the dates on wooden parts of some metal items of a hoard found in the Huelva estuary ~3080–2790 cal BP (Ruiz-Gálvez, 1995b). In addition, the material culture does not offer diagnostic typological criteria with which to attribute assemblages to phases. Radiocarbon-dated LBA settlements in Southwest Iberia are few as well: El Trastejón, Setefilla, Monte Berrueco, and the mining camp of Chinflón (Pellicer and Hurtado, 1980; Rothenberg and Blanco, 1980; Aubet et al., 1983; Escacena and De Frutos, 1985; Mederos, 1996; Hurtado et al., 2011). This knowledge base is changing only slowly (e.g. contributions in Celestino-Pérez and Rodríguez, 2017).

During the LBA and the Early Iron Age (EIA), a major mining and metallurgical area can be identified in the Huelva region, in connection with copper and silver extraction in the Iberian Pyrite Belt and the presence of a natural harbour in the Huelva estuary (Hunt, 2003). Around ~3200/3100 cal BP, tin-bronze items were ubiquitous in the study area; furthermore lead started circulating, both for producing ternary alloys (Cu, Sn, and Pb) and for extracting noble metals from argentiferous galena. By then, local Bronze Age communities had practiced relatively simple, rudimentary metallurgical techniques to produce copper-based items and to benefit native silver ores, especially during the EBA (Hunt, 2003; Nocete et al., 2008, 2011; Nocete, 2014). Although native silver is scarce in the region, argentiferous sulfurs proliferate and eastern Mediterranean know-how allowed the extraction of silver from them. Cupellation – a method for extracting silver from argentiferous jarosite adding lead – started being used around this time, likely via Phoenician craftmanship (Murillo-Barroso et al., 2016). At this juncture, the local abundance of argentiferous raw materials, the awareness of technology for extracting silver, and the circulation and availability of lead from elsewhere – most likely from southeast Iberia – were crucial circumstances in boosting the scale and volume of local metallurgical activities. All in all, these developments made a difference with previous metallurgical activities in the earlier Bronze Age (Hunt, 2003; Ruiz-Mata and Gómez-Toscano, 2008; Murillo-Barroso et al., 2016) and had traceable effects in the landscape and social organization of LBA and EIA communities. In contrast to the hotly debated matter of pollution and deforestation during the Copper Age and the EBA in Southern Iberia, there is relative consensus on the likely impact of intensive mining and metallurgical activities in the LBA and especially during the EIA in the region (e.g. García-Alix et al., 2013). Extensive deforestation detected in the S1 record ~3140 to 2800 cal BP might be attributed to social practices related to these intensive activities in the Huelva region (Stevenson and Harrison, 1992); S1 registers maximum values of CHAR microcharcoals ~2965 cal BP (167×10 particles $cm^{-2} year^{-1}$) and ~2865–2830 cal BP ($119–223 \times 10$ particles $cm^{-2} year^{-1}$), as well as a remarkable decrease in deciduous *Quercus* values (< 1%) (Figs. 3 and 5).

It is probable that the gaining importance of mining and metallurgy had a severe environmental effect across the Aracena (range) landscape during the LBA; this is suggested by the pollen record from El Trastejón (García Sanjuán, 1999; Hurtado et al., 2011), which shows a highly deforested and anthropized landscape – high values of nitrophilous

pollen taxa –, with a mixed economy of arable agriculture (presence of pollen of cereals and *Vicia faba*) and livestock farming (high percentages of anthropozoogenous pollen taxa) (Martín-Consuegra and Ubera, 2011). This trend is also visible in the woodlands of the DNP, judging by the S1 pollen record (Fig. 5).

The lifestyles, settlement forms, and even the effective occupation and permanence of local communities during the LBA in the study area are the subject of conflicting points of view. LBA habitats and domestic quarters in Southwest Iberia prior to the Phoenician arrival ~2800 cal BP are scarce and poorly documented. The absence of diagnostic features in the material culture hinders typological dating (Belén et al., 1992; Belén and Escacena, 1995; Ruiz-Gil and López-Amador, 2001; Torres-Ortiz, 2002; Suárez and Márquez, 2014; Ferrer, 2017). Around 3400 cal BP, small coastal outposts were founded on promontories and natural shelters such as at Tarifa, Huelva, and El Carambolo (Fig. 7a). They are regarded as nautical stopovers related to long-distance exchange with the earliest central and eastern Mediterranean seafarers (Delgado, 2013). Further inland, LBA villages were often founded on hills and constructed with stone-walls (Fig. 7a), such as at Los Castrejones and El Castillo on the Agrio River (Aznalcóllar) or Niebla on the Tinto River (Hunt, 2003; Campos et al., 2006; Gómez-Toscano, 2006, 2009; Ruiz-Mata and Gómez-Toscano, 2008). Further inland, sites such as Pocito Chico, Acinipo, and Montemolín (Suárez and Márquez, 2014) comprise a few huts with oval plants, stone foundations, and earthen walls lacking inner divisions, looking like previous EBA huts such as those at El Trastejón (Hurtado et al., 2011; Suárez and Márquez, 2014).

Some authors have argued for a settlement hiatus in Southwest Iberia during the final LBA (~3200–2800 cal BP), just before the arrival of the Phoenicians ~2800 cal BP (e.g. Belén et al., 1992; Belén and Escacena, 1995; García Sanjuán and Odriozola, 2012; Escacena, 2014). Thus, for these authors a weak LBA demography would have been followed by a sharp population growth in the EIA. Others, by contrast, have argued against any such population decline during the local LBA, claiming that this is a misleading image and reflects insufficient fieldwork (e.g. Gómez-Toscano, 2006, 2008; Gómez-Toscano and Campos, 2008; Gómez-Toscano and Fundoni, 2010–2011; Gómez-Toscano et al., 2014). Gómez-Toscano and Campos (2008) have posited an early demographic increase and economic intensification in the region already in the LBA ~3200–2800 cal BP, before the arrival of the Phoenicians. For them, a sufficiently complex political and economic organization was necessary to attract foreign trade (Ruiz-Gálvez, 2013). This involved the organization of the territory to obtain specific resources, such as minerals and metal bearing ores in Aznalcóllar, an expanding agriculture regime that included vine-growing areas, and relatively complex farming systems (Vera and Echevarría, 2013). It also entailed the control of communications, both terrestrial routes and the outpost of Huelva, where Atlantic and Mediterranean products, ideas, and techniques circulated and were exchanged during the latest Bronze Age, before the Phoenician diaspora in the early 3rd millennium cal BP (Ruiz-Gálvez, 1995a, 2013; Gómez-Toscano and Fundoni, 2010–2011; Gómez-Toscano et al., 2014).

The paleoenvironmental record from the S1 core is highly significant in this regard (Figs. 3–5). In our view, the S1 pollen record demonstrates, without ambiguity, that human impact in the particular local setting of the DNP fluctuated and was very uneven from a diachronic approach. For the first phase of the LBA (~3500–3140 cal BP; LPAZ-6 to LPAZ-8), no palynological evidence of anthropization is detected: cereal pollen is absent, anthropogenic nitrophilous and anthropozoogenous pollen taxa values are very low and coprophilous fungi values are negligible. Thus, only sporadic evidence of human and livestock presence can be inferred from such weak signatures. For the second phase of the LBA (~3140–2800 cal BP; LPAZ-9), on the contrary, evidence of the human footprint in the DNP is unquestionable, as witnessed by the percentage increase in anthropogenic nitrophilous herbs, anthropozoogenous taxa, and coprophilous fungi, yet also by a substantial increase in CHAR values and the reappearance of cereal pollen. Therefore, judging by the S1 pollen record, there was more intensive use of the local environment and a clear return of human populations to the Doñana marshes from 3140 cal BP onwards, the last centuries of the LBA.

It can be hypothesised that the two EWEs registered in the Doñana marshland during the LBA, ~3500–3435 cal BP (EWE-3) and ~3185–3140 cal BP (EWE-4), reduced settlement to a very limited number of locations, barely traceable in the palaeoenvironmental record (e.g. the "Bronze espagnol II" at the marsh of Rajaldabas site). This hypothesis is even probable if we take into account that in the period between both EWEs – ~3435–3185 cal BP – the S1 pollen record does not show any evidence of human activity. After EWE-4, during the pre-Phoenician Late Bronze Age, by contrast, the marshland ecosystems of Doñana began to suffer again from the impact of anthropogenic activities (agriculture, livestock, fires, deforestation); people might have resettled this marshland setting again, after some centuries of isolation.

6. Conclusions

A multi-proxy approach has proved a powerful tool to identify environmental changes ~5000–2800 cal BP in the present-day marshes of the Doñana National Park. Major changes in vegetation composition and structure of the marshland ecosystems couple with paleoestuary configuration, climate variability, and human dynamics. The low marsh and both freshwater-submerged and floating aquatic macrophyte communities dominated during the humid periods, when the Guadalquivir paleoestuary harboured a brackish lagoon. In contrast, in the arid periods (e.g. beginning from 4.2 kyr cal BP) the high marsh was the dominant vegetation and more salt – as a consequence of an EWE – concentrated in the ground. The amplitude of the vegetation response shows the elasticity and resilience of marshland communities to changes in the morpho-sedimentary environment. In any case, the aquatic habitats of the Doñana marshland are enormously heterogeneous and fluctuating, depending on the marine influence, the tides, and the Holocene evolution of the paleoestuary. For all these reasons, it must be understood that the marshland ecosystem is a mosaic of plant communities in which species with very different ecologies can coexist.

Our study is the first to be carried out at high-resolution in the Doñana National Park, relating vegetation history, cultural dynamics, and human impact with climatic variability and the potential effects of extreme wave events on settlement patterns during the Copper and Bronze Ages. In this sense, the paleoenvironmental record at the S1 point, in the middle of the present-day low marsh of Hinojos, has proved to be sufficiently sensitive to detect the strategies and palaeoeconomic activities of the prehistoric populations living in the paleoestuary. During the Chalcolithic, ~5000–4200 cal BP, in addition to potential fishing and shellfishing activities – not yet well researched –, settlements in the DNP area relied on important arable and pastoral activities, which are recorded in the S1 pollen record by an increase in both anthropogenic nitrophilous and anthropozoogenous herbs and coprophilous fungi. This evidence is clearly at variance with the prevailing understanding documented in the archaeological literature regarding the genesis of the present-day marshland in the paleoestuary; according to these archaeological sources, the paleoestuary was nothing but a slowly-evolving large gulf from the Postglacial Maximum to the Roman period. By contrast, during the Early Bronze Age (~4200–3500 cal BP) and the first phase of the Late Bronze Age (~3500–3140 cal BP), arable activity and livestock husbandry activities ceased, as evidence of anthropization is practically non-existent in the S1 pollen record for these periods. It is likely that the impact of three EWEs over such a time span (~4200–4000, 3500–3435 and 3185–3140 cal BP) limited not only the number of settlements in the DNP area and its vicinity but also their possible economic activities. These observations allow us to hypothesize the effective depopulation – or the ephemeral occupation by small scattered groups – of the study

area during both phases. Later, during the pre-Phoenician Late Bronze Age (~3140–2800 cal BP), the study area was resettled and the impact of human subsistence practices (agriculture, livestock raising, fires, deforestation) left recognisable traces on vegetation formations in a context of increasing climatic aridification. Overall, this account serves to enhance the cyclical, punctuated nature of local settlement dynamics during the studied timeframe.

Geomorphological, palaeobiological, and sedimentological features generated by EWEs during the Holocene are common along the coasts of the Gulf of Cadiz, having been assigned to either tsunamis or storm surges. However, except for a few examples, palaeoenvironmental data are not always conclusive enough to determine without ambiguity whether tsunamis or storms-surges were the real cause of such features. It would be advisable to extend the detailed study of the record of EWEs to determine their true origin, as well as to accurately establish their cause-effect relationship on prehistoric and historic settlement models and on the evolution followed by marshland ecosystems. Similarly, in order to adequately assess both vegetation dynamics and prehistoric occupation of these territories, more high-resolution palaeoenvironmental studies are needed, since the vast majority of those carried out so far have failed to meet this methodological approach.

Finally, attention should also be given to both the calibration of radiocarbon dates obtained in these marshland contexts in Southwest Iberia and the correction of the reservoir effect on samples from marine shells. Correction of this effect not only depends on the geographical location of the samples to date but also of its uncalibrated age. It is important to point out that in many cases bulk sediment has been dated without correcting for the reservoir effect. Such an oversight can result in a significant methodological error, since the sediments of the Doñana marshland usually contain millimeter fragments of marine shells and degraded remains of aquatic plants that are also a source of the hard-water effect, the magnitude of which has not been measured yet.

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